U.S. DEPARTMENT OF COMMERCE
National Technical Information Service
PB80-112162

Systematic Sensitivity Analysis of Air Quality Simulation Models

Science Applications, Inc, Pleasanton, CA

Prepared for

Environmental Sciences Research Lab, Research Triangle Park, NC

May 79

United States
Environmental Protection
Agency

Environmental Sciences Research
Laboratory
Research Triangle Park NC 27711

EPA-600/4-79-035
May 1979

Research and Development

PB80-112162

EPA

Systematic Sensitivity Analysis of Air Quality Simulation Models

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U. S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA. 22161

RESEARCH REPORTING SERIES

Research reports of the Office of Research and Development, U.S. Environmental Protection Agency, have been grouped into nine series. These nine broad categories were established to facilitate further development and application of environmental technology. Elimination of traditional grouping was consciously planned to foster technology transfer and a maximum interface in related fields. The nine series are:

1. Environmental Health Effects Research
2. Environmental Protection Technology
3. Ecological Research
4. Environmental Monitoring
5. Socioeconomic Environmental Studies
6. Scientific and Technical Assessment Reports (STAR)
7. Interagency Energy-Environment Research and Development
8. "Special" Reports
9. Miscellaneous Reports

This report has been assigned to the ENVIRONMENTAL MONITORING series. This series describes research conducted to develop new or improved methods and instrumentation for the identification and quantification of environmental pollutants at the lowest conceivably significant concentrations. It also includes studies to determine the ambient concentrations of pollutants in the environment and/or the variance of pollutants as a function of time or meteorological factors.

This document is available to the public through the National Technical Information Service, Springfield, Virginia 22161.

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. RECIPIENT'S ACCESSION NO.
EPA-600/4-79-035		PB80-112162

4. TITLE AND SUBTITLE	5. REPORT DATE
SYSTEMATIC SENSITIVITY ANALYSIS OF AIR QUALITY SIMULATION MODELS	May 1979
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
R. J. Gelinas and J. P. Vajk	
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
Science Applications, Inc. 80 Mission Drive Pleasanton, CA 94566	1AA603A AB-042 (FY-79)
	11. CONTRACT/GRANT NO.
	68-02-2942
12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Environmental Sciences Research Laboratories-RTP, NC Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, N.C. 27711	Final 1/78 - 12/78
	14. SPONSORING AGENCY CODE
	EPA/600/09

15. SUPPLEMENTARY NOTES

16. ABSTRACT

Systematic sensitivity and uncertainty analysis methods are reviewed and assessed for applications to air quality simulation models. The candidate methods in terms of their basic variables, mathematical foundations, user motivations and preferences, computer implementation properties, and costs (including both human and computer resources) are discussed. Both deterministic and sampling methods have been evaluated with illustrative examples of "What you pay" and "What you get" with present generation systematic sensitivity analysis methods and air quality models. Deterministic methods include the time- and user-honored methods of arbitrary parameter adjustments by trial and error, variational methods, and a newly formulated Green's function method (for kinetics systems only). Sampling methods include Monte Carlo sampling of the outputs of air quality models to compute variances and a Fourier analysis method of sampling model outputs to compute expectation values of sensitivity coefficients.

Computational economics, inclusive of both programming effort and computer execution costs, were found to be dominant governing factors for the effective application of systematic sensitivity and uncertainty analysis methods to arbitrary air quality problem scenarios; several reasonable options and several unreasonable options emerge from the present evaluations. Recommendations are made outlining how EPA should, today, most effectively apply available multi-parameter systematic sensitivity analysis methods to arbitrary 3-D transient (PDE) air quality simulation models.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
* Air pollution * Mathematical models * Analyzing * Probability theory		13 B 12 A 14 B

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSIFIED	107
	20. SECURITY CLASS (This page)	22. PRICE
	UNCLASSIFIED	A06 - A01

EPA Form 2220-1 (9-73)

EPA-600/4-79-035
May 1979

SYSTEMATIC SENSITIVITY ANALYSIS OF AIR QUALITY SIMULATION MODELS

by

Robert J. Gelinas and J. Peter Vajk

Science Applications, Inc.
1811 Santa Rita Road, Suite 104
Pleasanton, California 94566

Contract No. 68-02-2942

Project Officer

Robert E. Eskridge
Meteorology and Assessment Division
Environmental Sciences Research Laboratory
Research Triangle Park, North Carolina 27711

ENVIRONMENTAL SCIENCES RESEARCH LABORATORY
OFFICE OF RESEARCH AND DEVELOPMENT
U.S. ENVIRONMENTAL PROTECTION AGENCY
RESEARCH TRIANGLE PARK, NORTH CAROLINA 27711

DISCLAIMER

This report has been reviewed by the Environmental Science Research Laboratory, U.S. Environmental Protection Agency, and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the U.S. Environmental Protection Agency, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

ABSTRACT

This report reviews and assesses systematic sensitivity and uncertainty analysis methods for applications to air quality simulation models. The discussion of the candidate methods presents their basic variables, mathematical foundations, user motivations and preferences, computer implementation properties, and costs (including both human and computer resources). Both deterministic and sampling methods have been evaluated with illustrative examples of "What you pay"and "What you get" with present-generation systematic sensitivity analysis methods and air quality models. Deterministic methods include the time- and user-honored method of arbitrary parameter adjustments by trial and error, variational methods, and a newly formulated Green's function method (for kinetics systems only). Sampling methods include Monte Carlo sampling of the outputs of air quality models to compute variances and a Fourier analysis method of sampling model outputs to compute expectation values of sensitivity coefficients.

Computational economics, inclusive of both programming effort and computer execution costs, are the dominant governing factors for the effective application of systematic sensitivity and uncertainty analysis methods to arbitrary air quality problem scenarios; several reasonable options and several unreasonable options emerge from the present evaluations. Recommendations outline how the EPA should, today, most effectively apply available multi-parameter systematic methods of sensitivity analysis to arbitrary 3-D transient (PDE) air quality simulation models. The report concludes with a discussion of key breakthroughs which would lead to the next major advances in systematic sensitivity analysis. In certain instances, these would simultaneously provide major advances in air quality simulation modeling, *per se*.

This report was submitted in fulfillment of Contract No. 68-02-2942 by Science Applications, Inc., under the sponsorship of the U.S. Environmental Protection Agency. This report covers a period from May 12, 1978, to November 12, 1978, and work was completed as of December 4, 1978.

CONTENTS

Abstract iii

Figures vi

Tables vii

Acknowledgments viii

1. Introduction 1

2. The Problem 6

Air Quality Models and Their Sources of Uncertainty . . 6

Motivations for Sensitivity Analysis 12

Analytical Basis of Sensitivity Analysis Methods 14

3. Systematic Sensitivity Analysis Methods 30

Deterministic Methods 30

Sampling Methods 48

4. Evaluation of Candidate Sensitivity Methods for Air Quality Model Applications 61

Basic Cases 63

Chemical Model Evaluation 66

1-D Time-Dependent Model Evaluation 76

3-D Air Quality Model Evaluation 82

5. Today's Approach 85

Conclusions 85

Today's Practical Application of Systematic Sensitivity Analysis 87

Needed Breakthroughs in Systematic Sensitivity Analysis Methods 92

References 95

Preceding page blank

FIGURES

Number		Page
1	Effects of parameter variations on the solutions	19
2	Schematic representation of solution surfaces	22
3	Interpretation of probability distributions of uncertain parameters	24
4	Construction of first-order sensitivity measures and coefficients	26
5	Schematic illustration of the Monte Carlo method	49
6a	Schematic illustration of the Fourier Amplitude Sensitivity Test (FAST).	52
6b	Functional output of FAST	54
7	Arbitrary nature of functional forms used in FAST	56

TABLES

Number		Page
1	Characteristics of candidate systematic sensitivity and uncertainty analysis methods	60
2	Base cases for illustration of execution costs of candidate sensitivity analysis methods	83
3	Illustrative computer execution costs for candidate systematic sensitivity analysis methods	84

ACKNOWLEDGEMENTS

It is a pleasure to thank Dr. Richard Stolarski and Prof. John Seinfeld for in-depth discussions of their sensitivity work prior to publication. We are also extremely grateful to our colleague, Dr. Said Doss, for his exacting review and suggestions regarding some of the complex issues in numerical analysis which were encountered in the course of this study.

SECTION 1

INTRODUCTION

Uncertain parameters and sparse field data are intrinsic elements of air quality modeling. This state of affairs will persist indefinitely, despite the nation's ever more urgent need for more detailed understanding of cause and effect mechanisms in air quality. Thus a key question is, "To what extent do uncertain input factors in air quality models lead to impaired accuracy and precision of the resultant outputs of these models?" This basic question and many of its variants are examined in this final report on "Systematic Sensitivity Analysis for Air Quality Simulation Models".

The principal objectives of this study were:

(1) to identify those systematic sensitivity analysis methods which are applicable to air quality simulation models;

(2) to evaluate critically the potential of candidate methods of systematic sensitivity analysis for effective application to key air quality issues;

(3) to identify those breakthroughs most critically needed for each candidate method; and

(4) to determine the most effective immediate means of conducting sensitivity analyses on existing air quality simulation models, assuming no further developments in sensitivity analysis methods.

It is frequently unrecognized that sensitivity analysis is a basic ingredient in any application of the classic scientific method in which alternative hypotheses--including models and input data--are tested and retested against selected physical realizations. Historically, this process has most frequently been conducted on a trial and error basis. In the development of air quality models, sensitivity testing is a particularly laborious process if all potentially significant input parameters are to be varied, individually or simultaneously, for every potentially significant spatial location.

Somewhat more systematic approaches to sensitivity analysis, including some semi-automated methods, have appeared recently in the study of purely chemical kinetics systems. Enormous benefits would undoubtedly be realized in air quality analysis and policy making if semi-automatic sensitivity methods could be extended to economic applications in arbitrary space- and time-dependent multivariate models. Such extensions of systematic sensitivity analysis thus represent both a challenge and a promise for the entire field of air quality work. The principal focus of this study is the evaluation of this duality --the challenge versus the promise--in the application of candidate sensitivity analysis methods to arbitrary air quality models.

Perhaps the key consideration for air quality applications is the computational economics of candidate sensitivity methods. On the one hand, we shall see that several dedicated methods would require that a specific sensitivity program be written for each specific application to an air quality model. On the other hand, we shall see that several non-dedicated methods can be applied, without additional programming effort for either the air quality model or the general sensitivity

method, to any air quality model. All sensitivity analysis methods considered--in their current state of development--will be seen to have both strengths and weaknesses in regard to potential applications to 1-D, 2-D, and 3-D time-dependent air quality models. The extent to which the weaknesses will be disabling and/ or the extent to which the strengths can be exploited will necessarily depend upon the needs and expectations of potential users of these systematic methods. The motivations of air quality policy makers and of air quality researchers, while closely coupled, are sufficiently distinct that they may be served best by different methods of viewing uncertainties and sensitivities.

In the present study, it has been necessary to emphasize issues relating to the sensitivities of air quality models with respect to those physical, chemical, or meteorological factors which are most readily quantifiable. Thus we have considered variations in model outputs--such as the concentrations of O_3, NO, NO_2, and other air-borne species--which result from uncertain and variable input factors--such as photochemical rates, emission sources, pollutant sinks, and winds. We have not considered sensitivities to computational factors since no practical standards exist for comparative analysis. It would be impossible, for example, to determine the dispersion in oxidant concentrations attributable to numerical diffusion and/or to operator-splitting in the Systems Applications, Inc., regional model[1] *vis-a-vis* the LIRAQ model[2] *vis a vis* the MADCAP model[3] for a specific "typical day" in, say, St. Louis, even if the same data bases, initial conditions, and boundary conditions were incorporated

in each model. The multitude of other uncontrolled differences among air quality models precludes meaningful consideration of model-to-model sensitivities at this time.

We have thus surveyed and evaluated those sensitivity analysis methods which can, for any given air quality model, determine systematically and quantitatively the dispersion in the output variables--such as space- and time-dependent concentrations of ozone, PAN's, NO_x, acids, aldehydes, peroxides, and SO_x-- which result from uncertain photochemical rate data, winds, diffusivitives, initial and boundary values, and emission sources and sinks, and from deviations of actual conditions from "typical day" conditions.

Four types of systematic sensitivity methods are discussed in this report:

(1) "time-honored methods," which examine variations of selected parameters, either individually or simultaneously, by trial and error;

(2) variational methods (including direct methods);

(3) Fourier amplitude sensitivity test (FAST) methods;

(4) Monte Carlo methods.

The report is presented in four sections. In Section 2, The Problem, we discuss the motivations, logical bases, and key issues involved in the application of systematic sensitivity analyses to air quality simulation models. In Section 3, Systematic Sensitivity Analysis Methods, we describe the candidate methods in terms of their basic variables, mathematical formulation, computer implementation, and execution properties.

In Section 4, Evaluation of Candidate Sensitivity Methods for Air Quality Model Applications, we discuss "what you pay","what you get", areas of promise, and problems. Section 5, Today's Approach, indicates the most effective means, in our opinion, of applying systematic sensitivity methods to air quality models in the face of the current adverse constraints of computer economics. Finally, we indicate some of the needed breakthroughs which would lead to dramatic advances in the practical utility of candidate sensitivity methods.

SECTION 2

THE PROBLEM

AIR QUALITY MODELS AND THEIR SOURCES OF UNCERTAINTY

Air quality simulation models attempt to describe the physical, chemical, and photochemical processes occurring in specific portions of the lower atmosphere. Variables of principal interest include the concentrations of suspended particulate matter and of such gas phase chemical species as ozone, nitrogen oxides, and carbon monoxide, for which regulatory standards have been set by various governmental jurisdictions. Although no such regulatory standards have been set for species such as aldehydes, peroxides, nitrates, and acids, these and other species are also the subject of extensive current interest. The most thoroughly developed and calibrated air quality models available today describe, for the most part, the photochemical kinetics and the transport processes for gas phase species in certain limited atmospheric regions.

Several dissimilar sources of uncertainty in our knowledge of atmospheric processes affect the predictions of air quality simulations. Such uncertainty factors include:

(i) Natural Variabilities

- winds
- solar flux
- humidity
- natural sources and sinks of air-borne chemical species
- temperature
- atmospheric turbulence
- atmospheric stability
- particulate matter

It is important to note here that significant natural variabilities occur in the real atmosphere over scales which are often disparate with respect to the scales used in practical computations. For tropospheric models, typical computational scales are tens of meters to tens or hundreds of kilometers. Mixing of chemical species, on the other hand, occurs on microscopic scales comparable to molecular mean free path lengths. Small scale eddies and chemical mixing are thus, in most instances, treated by sub-grid scale characterizations rather than by detailed calculations of reactive turbulence. Winds at specific grid points in air quality models are generally interpolated from discrete field data from monitoring stations separated far wider in space and in time than the horizontal or vertical grids or integration time-steps of the models.

Variable attenuation and multiple scattering effects associated with particulate matter, gaseous components, cloud cover, and surface albedo all contribute to significant local variations in solar flux. Coastal areas, in particular, show extreme variations within a single geographic area and within a single day.

Naturally occurring chemical sources include forest fires, ocean spray, dust, local geogenic emissions from oilfields and coalfields, eutrophication of inland waters and oceanic processes releasing hydrocarbons, N_2O, CO, NH_3, and H_2S, as well as a nearly universal background release of methane. To these are added widespread sources attributable to agriculture and deforestation around the globe. Natural chemical sinks include hetero-

geneous reactions with suspended particulate matter, rainout, absorption at the air/ocean interface, and other surface deposition processes.

(ii) Photochemical Factors

- upper and lower atmospheric compositions (are determinants of solar flux intensity and spectral distribution)
- diurnal variations in solar flux
- absorption and photodissociation cross-section data
- hydrocarbon emission inventories
- photochemical characterizations of aggregate hydrocarbon emissions*

Photodissociation rates are described by wavelength integrations over products of local solar flux and photodissociation cross-sections. These rates thus depend strongly on the spectrum of the local solar flux which is determined by atmospheric composition and time of day[6]. Because the concentrations of minor reactive species--such as OH, O, H_2O_2, NO_3, and N_2O_5--have seldom been measured simultaneously with solar flux and the concentrations of other important atmospheric species, much of the validation of photochemical mechanisms remains to be done.

Atmospheric hydrocarbon inventories, of course, consist of huge numbers of individual organic compounds which must be aggregated according to their reactive properties in a photochemical mechanism. Additionally,

*See, for example, the articles of Hecht, Seinfeld, and Dodge(4), Hogo and Whitten (5), and Gelinas and Skewes-Cox(6) for detailed characterizations and categorizations of reactive hydrocarbons.

detailed emission inventories and local hydrocarbon distributions are difficult and costly to obtain for regions as extended as those spanned by air quality simulation models.

(iii) Field Data

Measurements of such meteorological variables as wind, temperature, stability, and transported chemical species are relatively sparse in both time and space. Because these quantities are associated with exceedingly complex fluid dynamics processes they have usually not been calculated from fundamental equations but have instead been imposed upon air quality simulation models using interpolations of the measured data onto the spatial grids and time points of the model. This imposition introduces uncertainties which are associated with the specific interpolation processes as well as uncertainties which are associated with non-representative measuring stations and with the measurement methods *per se*. An additional form of uncertainty arises because meteorological conditions are necessarily categorized to represent conditions which occur on selected "typical days". Actual meteorological conditions on any specific day will naturally differ from these "typical day" conditions, thus constituting yet another possible source of dispersion in the outputs of air quality simulation models.

(iv) Computational Factors

Air quality simulation models are formulated from non-linearly coupled partial differential equations in space and time describing physical transport and chemical kinetics processes. Very simple models are sometimes derived from systems of non-linearly coupled ordinary differential equations in time only. In either case, numerical integration methods are the most effective means for obtaining solutions of the basic modeling equations. Such numerical solutions are inherently approximate, so that simulation model results inevitably include dispersion due to these computational (numerical) approximations.

Computational dispersion usually arises from the well-known dilemmas of numerical modeling, most notably numerical diffusion (in Eulerian formulations) and aliasing of individual species under rezoning (in Lagrangian formulations). Such computational features as choice of differencing methods, representations of sub-grid scale physics, simplifications in chemical kinetics mechanisms, and imposed profiles for pollutant sources result in additional computational dispersions in the model outputs.

Model descriptions of the processes mentioned above are incorporated in systems of partial differential equations (PDE's) which are generally solved by numerical integration on large digital computers. In the present study, we considered air quality simulation models based upon PDE's of the form

$$\frac{\partial c_i}{\partial t} + \underline{\nabla}\cdot\left[c_i\underline{v} - \underline{\underline{K}}_D\cdot\underline{\nabla}c_i\right] = R_i(c_1,c_2,\ldots,c_N) + S_i(\underline{x},t), \quad i = 1,\ldots N, \quad (2.1)$$

where $c_i \equiv c_i(\underline{x},t)$ represents the mean concentration of the i-th chemical species; $\underline{v}$ is the mean fluid velocity; $\underline{\underline{K}}_D$ is a diffusion tensor; R_i is the net sum of all chemical reaction rates for production or destruction of the i-th species; and $S_i(\underline{x},t)$ represents the net sum of all sources and sinks for the i-th species. This equation is a reduced version of more fundamental (microscopically exact) fluid dynamics equations, and thus incorporates well-known approximations and assumptions which are peripheral to the present discussion.

The solutions $c_i(\underline{x},\{p\},t)$ of Eq. (2.1) are functions of space and time ($\underline{x}$, t), but they are also functions of the parameters $\{p\} \equiv \{\underline{v}, \underline{\underline{K}}_D, k_\alpha, J_\beta, \mathcal{L}_\gamma, c_{i,I}, c_{i,B}\}$. The parameters $k_\alpha(T)$ represent temperature-dependent rate constants determining the thermal chemical reaction rates; J_β represents coefficients for the photochemical reactions included in the terms R_i; and $\mathcal{L}_\gamma$ represents parameters in the expressions for source and sink rates included in the terms S_i. Since the solutions for the species concentrations are functionally dependent on the initial and boundary values, $c_{i,I}$ and $c_{i,B}$, respectively, it is often useful to include these values in the parameter array $\{p\}$ as well.

The values for this host of parameters needed to describe important atmospheric processes are obtained from laboratory experiments, field measurements, and theoretical or empirical prescriptions. The source of the entire problem under discussion in this study is the inevitable fact that all of these parameter evaluations are uncertain to a degree. In the face of such uncertainties, it is natural to ask, "For which of the parameters do the uncertainties in the parameters produce the greatest dispersion in the model outputs (especially for the most important atmospheric observables)?"

MOTIVATIONS FOR SENSITIVITY ANALYSIS

Air Quality Policy

Air pollution control agencies and other governmental bodies or officers are charged with the responsibility of regulating pollutant emissions in order to meet or to maintain standards of air quality established for certain atmospheric regions. To the extent that models are used responsibly in decision making, it is both fair and necessary to ask, "What are appropriate error bars for the outputs of the model?" This question is appropriate whether the models used are analytically based, detailed simulation models incorporating as much of the current scientific knowledge as is practical or the models are qualitative mental models which incorporate intuition. In asking this question, we would like to obtain a statement of the uncertainty--or, equivalently, the credibility--of the model results which are used in regulatory decisions. The principal emphasis is then given to determining the expected uncertainties in important atmospheric variables, given the manifold of uncertainties present in the input parameters of

the model. (Analytic definitions will appear in later sections of this report.) As an example, one might want to know the expected uncertainty in ozone concentrations predicted by a given simulation model at all significant geographical locations and times resulting from the combined effects of rate data uncertainties, meteorological uncertainties, emission uncertainties, etc., in order to assess whether proposed regulations are reasonable and proper.

Air Quality Research

In contrast to regulatory policy applications of air quality simulation models in which (expected) uncertainty analysis--or credibility analysis--is emphasized, research applications tend to emphasize sensitivity--cause-and-effect-analysis. The two concepts are, to a significant degree, complementary. In research applications, the scientist seeks to determine the magnitude of the effects which each and every parameter input to the model produces in the outputs of that model. This is a more explicitly detailed approach than that of expected uncertainty analysis because, in any specific simulation calculation, the model must use a single, specific numerical value for each parameter of the model, not a distribution of possible values. Best case and worst case air quality estimates are very conveniently made in terms of individual, unaveraged parameter variations.

In studying the effects of varying an individual parameter (or a limited set of parameters) on the outputs of the model, all other model parameters are generally held fixed,

either at nominal "best" values or at "test values" selected by the researcher. (It is also possible to consider simultaneous variations in all other parameters in order to obtain best or worst case estimates.) Sensitivity analysis is thus used by researchers as a means for identification of cause-and-effect mechanisms and for ranking various mechanisms in order of importance in the overall system of processes determining the state of a specific atmospheric region. The goals of this approach are: (i) to increase basic scientific understanding of atmospheric processes; (ii) to focus scientific research on the most critical issues, ultimately reducing uncertainties in existing models; and (iii) to assist regulatory applications, both directly and indirectly, by progress toward the first two goals

ANALYTICAL BASIS OF SENSITIVITY ANALYSIS METHODS

The methods and language of statistics have been extensively adopted in the development of systematic sensitivity methods, undoubtedly because interest centers on deviations in model solutions which result from deviations in governing input parameters. In terms of air quality modeling equations which are represented by Eq. (2.1), a major question is, "What variations occur in the solutions, $c_i(\underline{x}, \{p\},t)$, relative to $c_i(\underline{x},\{p^o\},t)$, when model input parameters, $\{p\}$, are subjected to variations relative to their nominal values $\{p^o\}$?" The input and internal parameters for Eq. (2.1) are written most generally in terms of their nominal values and deviations as:

$$c_{i,I} \equiv c_{i,I}^{o} + \delta c_{i,I}$$

$$c_{i,B} \equiv c_{i,B}^{o} + \delta c_{i,B}$$

$$\underline{v} \equiv \underline{v}^{o} + \delta \underline{v}$$

$$\underline{\underline{K}}_D \equiv \underline{\underline{K}}_D^{o} + \delta \underline{\underline{K}}_D$$

$$k_\alpha \equiv k_\alpha^{o} + \delta k_\alpha$$

$$J_\beta \equiv J_\beta^{o} + \delta J_\beta$$

$$\mathcal{L}_\gamma \equiv \mathcal{L}_\gamma^{o} + \delta \mathcal{L}_\gamma \quad . \tag{2.2}$$

At this stage nominal values, $\{p^o\}$, are not necessarily mean values, nor are deviations, $\{\delta p\}$, necessarily standard or random deviations. Considerable care must thus be exercised to distinguish nominal values from mean values or central values, and also to distinguish arbitrary deviations from standard deviations, variances, and random variations.

The logical question of, "How are nominal values to be assigned in air quality models?", has several answers. In principle, measurable parameters such as reaction rate data, initial and boundary values, winds, diffusivities, etc. can be represented ultimately by mean values and associated standard deviations; it is frequently true, in practice, that available data bases do not allow reliable statistical treatment. For example, in photochemical kinetics some limited number of rate coefficients have been repeatedly measured by independent experiments which have been well-prepared (in the statistical mechanical sense) and thus

have reliable statistical properties. But this tends to be the exception rather than the rule for many reactions which are objects of current research. Indeed, we can not now profess to know all of the potentially important atmospheric reaction processes, much less their mean rates and statistical distributions. In such cases, nominal values of known and hypothetical reaction rates are assigned on the basis of each individual researcher's discretion. Parametric deviations are also arbitrarily assigned for the purpose of testing the sensitivity of photochemical mechanisms to specific reactions. The history of atmospheric photochemistry developments emphatically illustrates that meaningful probability distributions of poorly known reaction rates are simply not available at times of greatest need. (Poorly known chemical rates have always been subject to very large, frequently unpredictable changes which emerge when markedly new or more carefully prepared experiments are performed.) This discussion of meaningful probability distributions for physical parameters is particularly pertinent to our consideration of those sensitivity and uncertainty analysis methods which use probability distributions.

Measures of Sensitivity and Uncertainty

Several measures are used in sensitivity and uncertanity analysis which should be distinguished at the outset.

The most elementary measure is a deviation (variation) in state variables such as species concentrations at specific coordinates $(\underline{x}', t')$,

$$\delta c_i(\underline{x}';\{p\},t') \equiv c_i(\underline{x}',\{p^0 + \delta p\},t') - c_i(\underline{x}',\{p^0\},t'), \quad (2.3)$$

in the neighborhood of nominal parameter values, $\{p^0\}$. (Recall that the parameters can be either random or non-random variables.)

By Taylor's theorem

$$c_i(\underline{x}', \{p^o + \delta p\}, t') = c_i(\underline{x}', \{p^o\}, t') + \sum_{k=1}^{M} Z_{i,k}\, \delta p_k + O((\max \delta p_k)^2), \quad i = 1,N;\ k = 1,M, \tag{2.4}$$

where the sensitivity coefficients are defined by

$$Z_{i,k} = \partial c_i / \partial p_k \quad , \quad i = 1,N;\ k = 1,M \ . \tag{2.5}$$

Sensitivity methods which neglect the terms $O((\max dp_k)^2)$ are referred to as first-order or linear methods. In space and time dependent models, which are described by partial differential equations (in contrast to ordinary differential equations for purely kinetics models or for steady state purely fluid dynamics models), species concentration variations associated with the k^{th} parameter variations are given by the functional derivatives,

$$\delta c_i(\underline{x}', t') = \int \frac{\partial c_i(\underline{x}', t')}{\partial p_k(\underline{x}, t')}\, \delta p_k(\underline{x}, t')\, d^3x \tag{2.6}$$

and

$$\delta c_i(\underline{x}', t') = \int \frac{\partial c_i(\underline{x}', t')}{\partial p_k(\underline{x}', t)}\, \delta p_k(\underline{x}', t)\, dt. \tag{2.7}$$

The sensitivity variable in Eq. (2.6) represents variations in c_i at $(\underline{x}', t')$ with respect to variations in p_k at locations $\underline{x}$ and time t'. Likewise, the sensitivity variable in Eq. (2.7) represents variations in c_i at $(\underline{x}', t')$ with respect to variations in p_k at times t and location $\underline{x}'$. These functional derivatives and the sensitivity coefficients, $Z_{i,k}$, form the basis of

variational methods which will be described in Section 3. These variables provide convenient measures of sensitivities and uncertainties when probability distributions are poorly defined and for "best case" and "worst case" air quality considerations, which were discussed above in connection with AIR QUALITY RESEARCH MOTIVATIONS.

The effects of parameter variations on the solution $c_i(\underline{x},t)$ are shown schematically in Figure 1. Two cases must be distinguished here. In Figure 1(a), the solution for the "best value" of all parameters is shown by the solid line. If some one of the parameters, say p_j, were changed from its "best value" p_j^o to $p_j^o + \delta p_j$, the solution curve would be different, although it would start from the same initial value, $c_{i,I}$, for c_i. Likewise, if the parameter were changed to $p_j^o - \delta p_j$, the solution curve would be different again. These altered solutions are shown by the dotted lines in Figure 1(a). If, on the other hand, we wanted to investigate the effects of changing the initial value $c_{i,I}$ itself, the results might look like the dotted line curves shown in Figure 1(b).

It is important to realize that it is not enough to consider the variations in the solutions c_i at a single time t'; we must consider the variations themselves as functions of time if we are to properly evaluate the sensitivity of the system to parameteric variations or uncertainties.

If the set of coupled differential equations describing the system is non-linear, not only the <u>value</u> of c_i at time t may in general be altered, but the <u>shape</u> of the solution curve itself may change. In certain extreme cases, changing some parameters p_s ($s \neq j$) by small amounts may result in

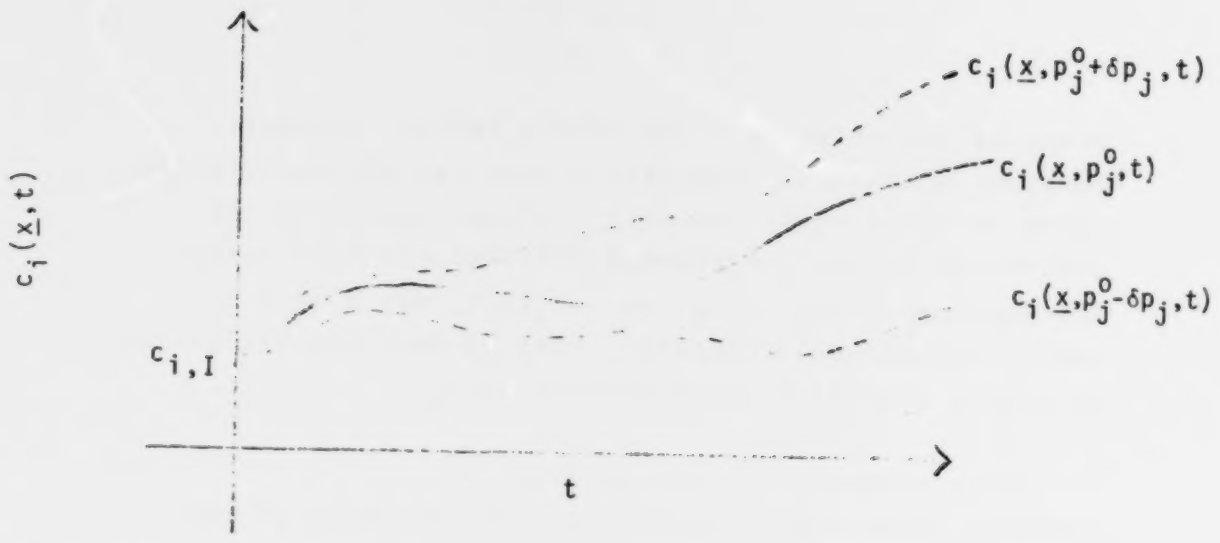

Figure 1(a). The effect on the solution for c_i as a function of time when one of the parameters p_j is altered by $\pm\ \delta p_j$, where p_j is one of the rate coefficients or boundary conditions. (For example, c_i may represent the concentration of ozone and p_j may represent a local photochemical rate coefficient.) The parameter p_j may also represent the initial value of a <u>different</u> concentration c_ℓ, $\ell \neq i$.

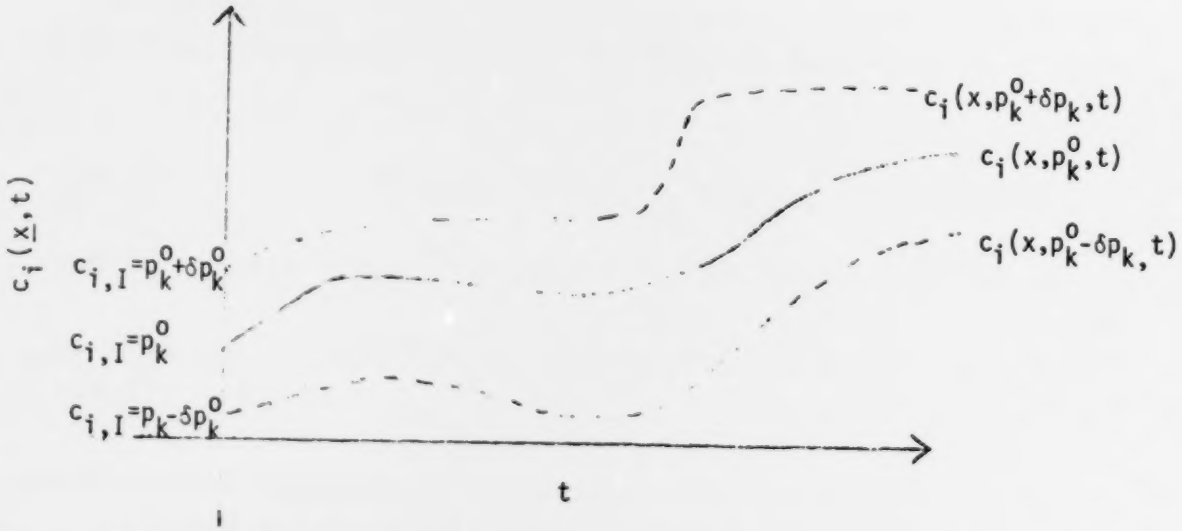

Figure 1(b). The effect on the solution for c_i as a function of time when the parameter p_k representing the initial value of c_i is altered by $\pm\ \delta p_k$. Because of the non-linearity of the system of coupled equations, the solution curves are <u>not</u> the same shape as the solution for the "best value" with a constant displacement. Changing the initial value can alter the qualitative shape of the concentration-versus-time curve significantly.

changing the solution curve from a smooth, gradually varying function of time into a very rapidly oscillating curve of small amplitude which follows generally the trajectory of the unperturbed solution. In such cases, the magnitude of the variation $\delta c_i(\underline{x},t)$ may always be small, but we would certainly want to describe the system as highly sensitive to variations in p_s.

Other measures are obtained by introducing statistical averages taken over distributions of parameter values. For example, mean species concentrations, $\langle c_i(\underline{x}',t')\rangle$, are defined by

$$\langle c_i(\underline{x}',t')\rangle \equiv \int\cdots\int c_i(\underline{x}';p_1\cdots,p_M;t')P(p_1,\ldots,p_M)dp_1\cdots dp_M\,, \quad i = 1,N, \qquad (2.8)$$

where $P(p_1,p_2,\ldots,p_M)$ represents the probability distribution for the parameters. Uncertainties in species concentrations are often expressed in terms of variances, $\sigma_i(\underline{x}',t')$, defined by

$$\sigma_i(\underline{x}',t') \equiv \langle \left[c_i(\underline{x}',t') - \langle c_i(\underline{x}',t')\rangle\right]^2\rangle \qquad (2.9a)$$

or

$$\sigma_i(\underline{x}',t') = \langle c_i(\underline{x}',t')^2\rangle - \langle c_i(\underline{x}',t')\rangle^2 \quad i = 1,N, \qquad (2.9b)$$

where

$$\langle c_i(\underline{x}',t')^2\rangle \equiv \int\cdots\int c_i(\underline{x}';p_1,\ldots p_M;t')^2\, P(p_1,\ldots,p_M)\, dp_1,\cdots dp_M, \quad i = 1,N \qquad (2.10)$$

Variances provide convenient measures of <u>expected</u> uncertainties, which were discussed above in connection with AIR QUALITY POLICY motivations. As for expectation values of sensitivity coefficients, sampling methods have been developed to evaluate measures of $\langle Z_{i,k}\rangle$, which will be described in Section 3.

GRAPHIC INTERPRETATION

A graphic interpretation of the preceding discussion may clarify the meanings of, and differences among, the various measures of sensitivity and uncertainty. For simplicity, suppose that the set of coupled differential equations (2.1) depends on only two uncertain parameters, p_1 and p_2, whose values lie between p_1^{min} and p_1^{max} and between p_2^{min} and p_2^{max}, respectively. For any given position and time $(\underline{x},t)$, the solution c_i can be considered to be a function of p_1 and p_2 as shown in Figure 2. The "best values" of p_1 and p_2 are, respectively, p_1^o and p_2^o; in general, these are not in the exact centers of the respective domains of uncertainty.

The point Q on the solution surface represents the magnitude of the solution c_i at time $(\underline{x},t)$ computed with both uncertain parameters assumed to have their "best values." If p_1 were then varied over its domain of uncertainty, holding p_2 fixed at its "best value," the solution point would trace out the curve DQD'. Conversely, if p_2 were varied over its domain of uncertainty, holding p_1 fixed at p_1^o, the solution point would trace the curve AQA'. Varying both of the parameters over the full domain of uncertainties in both then generates a two-dimensional solution surface for each variable c_i for each position $\underline{x}$. This solution surface changes as t itself is allowed to vary.

In the general case, far more than two uncertain parameters are involved in an air quality model. If M parameters are uncertain, the solutions trace out M-dimensional hypersurfaces; the features of the analysis discussed below apply equally well to the M-dimensional case as to the two-dimensional case.

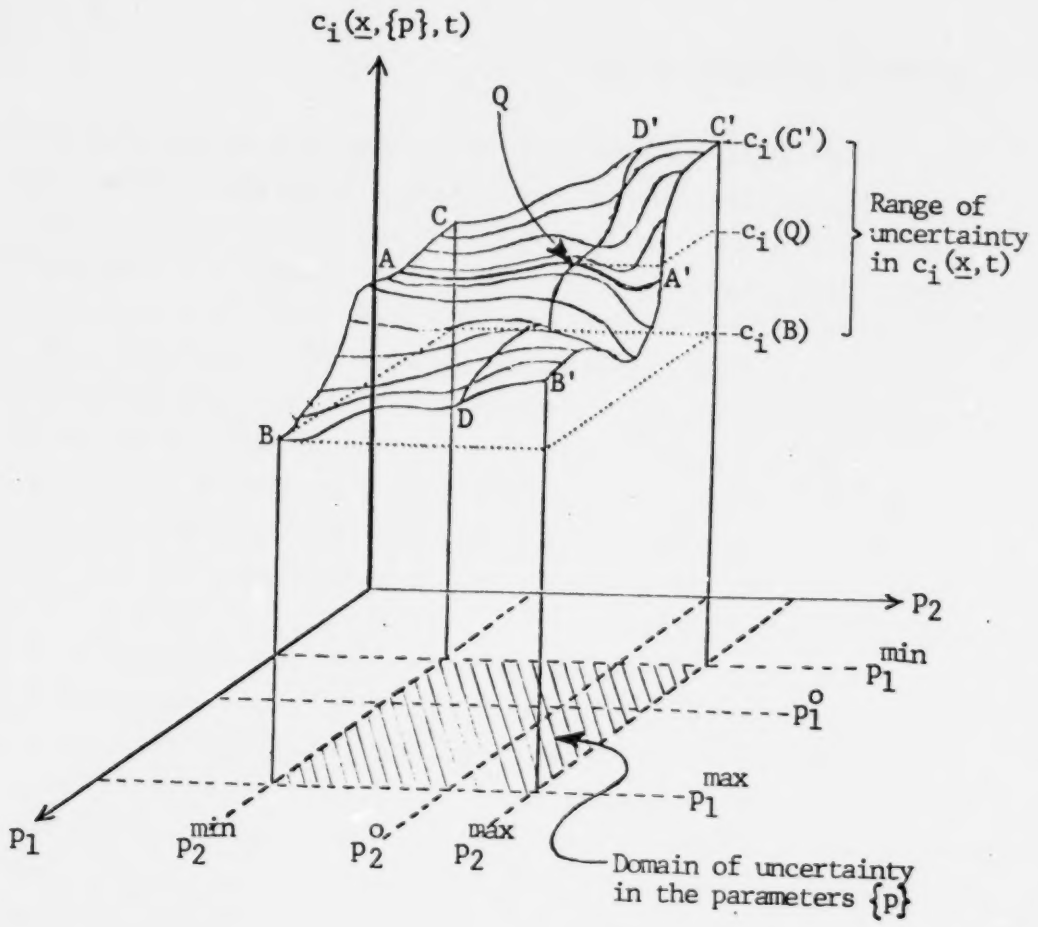

Figure 2. Schematic representation of the solution surface c_i over the domain of uncertainty in the parameters. The resulting range of uncertainty in the value of the solution c_i about the nominal value corresponding to the "best values" of the parameters is also indicated.

The solution surfaces $c_i(\{p\})$ may have very complicated topographies, with peaks, pits, undulations, sharp ridges or gorges, saddles, and even such discontinuities as sheer cliffs. The purpose of sensitivity analysis, from this point of view, is to understand the shape of the solution surfaces as we vary the uncertain parameters away from the immediate vicinity of the current "best values." As new field data or new experimental evaluations of parameters become available, our estimation of the "best values" may change, moving us across the complex topography of the solution surface. Understanding the shapes of the solution surfaces provides advance warning about what kinds of changes in the parameters will most drastically change the predictions of our simulation models.

As we mentioned earlier, for policy-making purposes uncertainty analysis is generally of greater interest than sensitivity analysis. The purpose of uncertainty analysis is to assay the range of uncertainty in the solutions c_i resulting from the present degree of uncertainty in the parameters, as indicated in Figure 2. A crude measure of this uncertainty is the difference in elevations between the nominal case (point Q) and the extremes (points B and C'). Although the extreme values of c_i are shown in the figure to lie at corners of the domain of uncertainty in the parameters, the extreme values will more commonly lie somewhere inside that domain.

If we have some idea about the probability distributions for the uncertain parameters, we can in principle compute the probability distribution for c_i over the domain of uncertainty in the parameters, as shown in Figure 3. From the probability distribution for c_i, quantities such as mean species concentrations $\langle c_i \rangle$ and variances σ_i can be computed

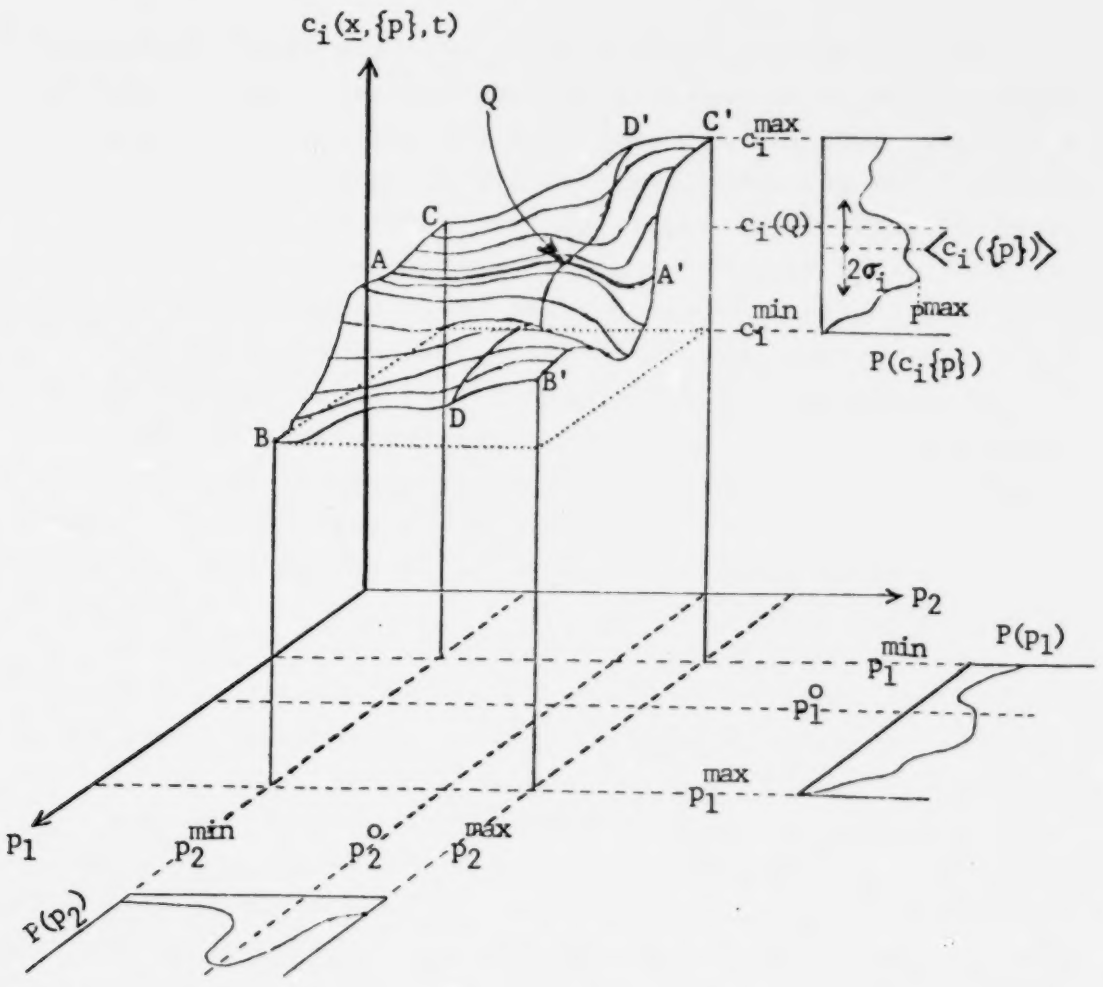

Figure 3. Given assumed probability distributions for each uncertain parameter, the probability distribution for the solution c_i is obtained by computing a probability-weighted distribution of elevations of the solution surface. Note that the "best value" of a parameter may differ, in general, from both the most likely value and the mean value of the parameter. Likewise, $\langle c_i\{p\}\rangle$, the mean value of the solution c_i, may differ from the most likely value of the solution and from the nominal value $c_i(Q)$ corresponding to the "best values" of the parameters. We can never be absolutely certain that the true value of a parameter lies within a given domain of uncertainty. Thus $P(p_1)$ and $P(p_2)$, in general, have small but finite values beyond the domain shown. In this illustration, the uncertainties in p_1 and p_2 are assumed to be independent and uncorrelated.

by standard statistical methods as defined in Eq's (2.8), 2.9), and (2.10).

Several important points should be noted in Figure 3. First, the investigator's estimate of the "best value" of a parameter may or may not coincide with either the most likely value or the expected (mean) value of the parameter as computed from the probability distribution. (Note, in particular, p_1 in the figure.) Second, the shape of the solution surface c_i is independent of all assumptions about the shapes of the probability distributions for the parameters; the shape of the solution surface depends only on the structure of the set of coupled differential equations (2.1). Third, the probability distribution for the value of the solution $c_i(\underline{x},t)$ depends on *both* the assumed probability distributions for the parameters *and* the shape of the solution surface. Thus the mean value $<c_i\{p\}>$ may or may not coincide with either the most likely value of c_i or the nominal value of c_i corresponding to the "best values" of the parameters. Fourth, the variance σ_i of the probability distribution for the solution c_i will in most cases be significantly smaller than the range of uncertainty in c_i, that is, $2\sigma_i \leq (c_i^{max} - c_i^{min})$.

The significance of Eqs. (2.3), (2.4), and (2.5) can be seen in Figure 4. If we make small changes in the parameters, one at a time, and compute c_i for the altered parameters, we find in general that c_i has changed by a small amount δc_i. Changing p_1 by a small amount δp_1 moves the solution point from Q along the curve DQD' toward D. (See figure.) Changing p_2 by a small amount δp_2 moves the solution point from Q along the curve AQA' toward A'. The differences in elevation resulting from these small displacements within the domain of uncertainty are given by Eq. (2.3).

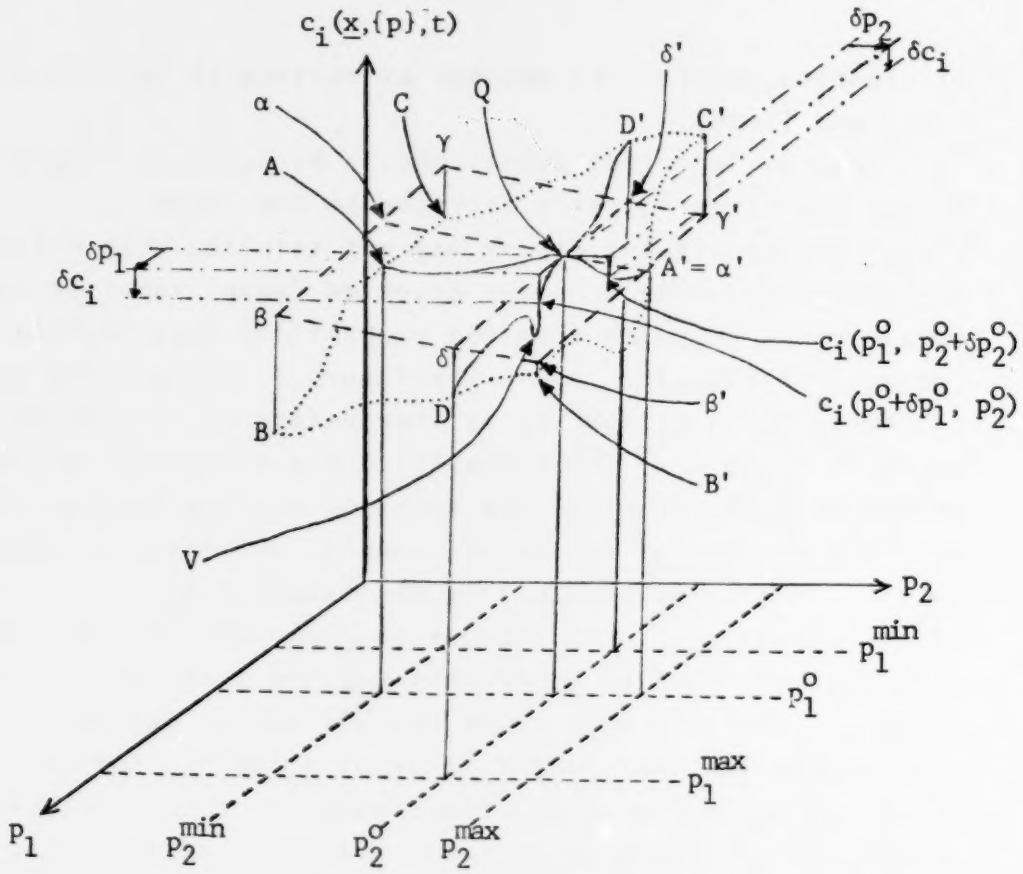

Figure 4. Construction of first-order sensitivity measures and coefficients. First-order sensitivity analysis approximates the true solution surface bounded by the curves BDB', BAC, B'A'C', and CD'C' by the tangent plane at point Q which is bounded by the straight line segments $\beta\delta\beta'$, $\beta\alpha\gamma$, $\beta'\alpha'\gamma'$, and $\gamma\delta'\gamma'$, respectively. Using this approximation severely underestimates the range of uncertainty in the solution c_i and provides no warning of such structural features as the deep valley at V.

In the limit of infinitesimal displacements δp_j, the ratio of the deviation in c_i to the displacement in p_j is the slope of the solution surface in the direction of the p_j axis at the point Q. The slopes thus defined are the first-order sensitivity coefficients $Z_{i,j}$ defined by Eq. (2.5). First-order sensitivity analysis, in effect, approximates the shape of the solution surface bounded by the curves BDB', BAC, B'A'C, and CD'C' in Figures 2, 3, and 4 by the plane bounded by the straight lines $\beta\delta\beta'$, $\beta\alpha\gamma'$, $\beta'\alpha'\gamma'$, and $\gamma\delta'\gamma'$ which is tangent to the solution surface at the nominal solution Q. (See Figure 4.)

For very small displacements about the "best values," the true solution surface differs from the tangent plane by only small differences in elevation. In this regime, comparison of first-order sensitivity coefficients tells us to which parameters the solutions are most sensitive. But first-order sensitivity analysis provides no warning whatever about such significant features as the deep valley at V, only a modest distance in parameter space from the "best values." Nor would this analysis indicate the full range of uncertainty in the solutions c_i over the domain of uncertainty of the parameters p . For the case shown in Figure 4, the range between extreme values on the tangent plane-- $\left[c_i(\gamma) - c_i(\beta')\right]$ -- is less than half the range for the true solution surface-- $\left[c_i(C') - c_i(B)\right]$. Moreover, the values of the parameters corresponding to the extreme values on the tangent plane are completely different from the values of the parameters for the true extremes of c_i.

If we wish to learn something about the shape of the true solution surface at finite distances from Q, we can perform a Taylor series expansion about $c_i(Q)$, evaluating not only the first derivatives of c_i with respect to all the parameters p_j, but also higher-order derivatives (higher-order sensitivity coefficients). Including second-

order coefficients would be an improvement over first-order sensitivity analysis, but it would be inadequate for the case shown in Figure 4 since Q is an inflection point of the curve DVQD'. Thus the second derivative $\partial^2 c_i / \partial p_1^2$ vanishes at Q, and we would still have no intimation of the valley at V.

In general, a truncated Taylor series approximation to the solution surface can be characterized by a *radius of approximate convergence*, a small distance in parameter space about the point Q within which the series approximation differs from the true solution by less than some specified ε. Regardless of how many terms are included in the expansion, however, the difference between the approximation and the true function depends on the still higher-order terms which have been omitted. Unless we have sound theoretical reasons to show that higher-order derivatives become vanishingly small (and this is almost never the case for realistic air quality models), we have no assurance that we will not overlook such features as the valley at V a very short distance away from Q. Consider, as an example, a function whose first and second derivatives are of order unity, while all higher derivatives vanish, except the 20^{th}, which has a value of 10^{100}. The terms involving this derivative are then of order $(10^{100})\ \delta p^{20}$, so that to achieve an $\varepsilon \sim 10^{-5}$, the radius of convergence would still be $\lesssim 10^{-6}$, even if we included all terms up through the 19^{th} order.

An alternative approach is to systematically examine the solution surface by computing c_i from the simulation model at a significant number of locations in the domain of uncertainty of the parameters $\{p\}$. Unless the functional forms involved in the various coupling, source, and sink terms in the set of differential equations (2.1) are simple expressions, it is difficult to be sure we have not accidentally overlooked

significant structures hidden between the locations actually examined. Several of the sampling methods discussed in a later section attempt to deal with this problem in one way or another.

We noted earlier that the shapes of the solution surfaces c_i ({p}) depend only on the structure of the basic set of coupled differential equations (2.1). If the functional forms for the sources, sinks, photochemical rates, or chemical kinetics rates are changed, or if new reactions are added to the system, or other reactions are removed from the system, all of the solution surfaces are changed. Peaks, pits, undulations, ridges, gorges, and cliffs in the surfaces corresponding to any particular chemical species c_k, for example, will all be altered; some may disappear, while new topographic features may appear. The shapes of the distribution functions $P(c_i)$ deduced for the values of the solutions c_i (see Figure 3) and the range of uncertainty for c_i (see Figure 2) will also be changed. Thus, <u>whatever</u> method of sensitivity analysis or uncertainty analysis is used, the evaluation of sensitivity or uncertainty must be repeated <u>ab initio</u> each time the system of equations (2.1) is altered.

SECTION 3

SYSTEMATIC SENSITIVITY ANALYSIS METHODS

Concise descriptions are given in this section of the candidate systematic sensitivity analysis methods, starting with the logically simplest "time-honored" method and proceeding next to variational methods and, finally, to sampling methods. We subdivide this section into two major subsections: Deterministic Methods and Sampling Methods. The deterministic methods include both the "time-honored" and the variational methods.

Arbitrary, unaveraged deviations, δc_i and $Z_{i,k}$, are the measures of sensitivity used in deterministic methods, whereas expectation values, $\sigma_i(\underline{x}', t')$ and $\langle Z_{ik} \rangle$, are the measures used in sampling analyses. Table 1 summarizes the salient features which appear in this section.

DETERMINISTIC METHODS

The Time-Honored Method

Trial-and-error testing of model parameters has stood the test of time in all scientific disciplines. This time-honored method continues to be the most widely used sensitivity testing method because it is flexibly applied to arbitrary problem applications and, most significantly, because most researchers understand the trial-and-error method and are comfortable with it. Developments in our present study suggest that the time-honored method will continue to have an important role to play in conjunction with applications

of more-highly automated sensitivity tests to complex air quality simulation models.

The time-honored sensitivity method is applied by simply altering the value of model parameter, p_j, at any $(\underline{x}, t)$ and then noting the deviations,

$$\delta c_i(\underline{x}, \{p\}, t) = c_i(\underline{x}, p_j^o + \delta p_j, t) - c_i(\underline{x}, p_j^o, t), \quad (3.1)$$

which are obtained from the respective model solutions for $c_i(p_j^o + \delta p_j)$ and $c_i(p_j^o)$. (See Figure 4.) The major features of this method are:

- Arbitrary deviations δc_i are economically obtained for applications having poorly defined probability distributions of model parameters.
- Variances $\sigma_i(\underline{x}', t')$ and expectations of sensitivity coefficients $\langle Z_{i,k} \rangle$, $\langle Z^{(2)}_{i,jk} \rangle$, etc., can be evaluated but may be extremely costly to compute. (Examples will be given in Section 4.)
- Sensitivities and uncertainties which are continuous in time, t, are obtained for the entire problem time domain.
- It is a non-dedicated method; that is, the time-honored method applies to arbitrary air quality simulation models without any need to alter the air quality simulation model itself, or to develop special programming for performing sensitivity analyses.

- Variations in photochemical kinetics and fluid dynamics parameters are treated with equal facility (whether the chemical kinetics and the fluid dynamics are treated separately or in fully coupled form) over the entire space and time domains of interest. Applications to date include the full range of coupled kinetics and hydrodynamics.

Clearly, the time-honored method has a tremendous combination of simultaneous features which other sensitivity methods do not yet have; and, depending upon the craft and specific motivations of the practicing scientist, the method is also both systematic and economical.

Variational Methods

As we discussed in Section 2 above, the basic task of sensitivity analysis is to understand the shapes of the solution surfaces defined by considering the solutions $c_i(\underline{x}, \{p\}, t)$ at any given position and time to be functions of the uncertain parameters of the system. If we consider such a solution surface (as was illustrated in Figures 2, 3, and 4) at a sequence of times, the surface itself changes shape with time. If we are interested in just a small neighborhood of the "best values" of the parameters $\{p\}$, we could in principal compute the solutions $c_i(\underline{x},t)$ for a large number of sets of values of $\{p\}$in the vicinity of the "best values" $\{p^o\}$ and then laboriously construct the solution surfaces for each time of interest to obtain numerical approximations to the sensitivity coefficients $Z_{i,k}$, $Z^{(2)}_{i,kl}$, etc., by straightforward but tedious means.

But a far simpler method is to treat the sensitivity coefficients themselves as dynamic variables. In other words, we want to obtain equations of motion describing the change in time of the slopes and curvatures, etc., of the solution surfaces. These can be derived analytically and then solved once by numerical methods instead of solving the original set of equations (2.1) many, many times for different sets of values of $\{p\}$ in the vicinity of $\{p^o\}$. Variational methods, in brief, are based on this latter approach.

(i) Chemical Kinetics with Constant Rate Coefficients

Most automated sensitivity analyses implemented thus far have been applied to the case of purely chemical kinetics in which rate coefficients are held constant in time. Variational methods are also sometimes referred to as direct methods when problem parameters are held constant. By any terminology, variational methods are computationally dedicated in the sense that a set of equations for sensitivity coefficients must be derived and solved in conjunction with the original model equations.

Photochemical kinetics model equations are nonlinearly coupled ordinary differential equations of the form

$$\dot{c}_i \equiv dc_i/dt = R_i(c_j,\{p\}, t), \qquad i,j = 1, N. \qquad (3.2)$$

In the absence of spatial dependences, the parameter array $\{p\}$ contains all of the variables listed in Eq.(2.2), except $c_{i,B}$, $\underline{v}$, and $\underline{\underline{K}}_D$.

The sensitivity coefficients are defined as partial derivatives of the solutions c_i with respect to the uncertain parameters:

$$Z_{i,k} \equiv \frac{\partial c_i}{\partial p_k} , \qquad i=1,N, \quad k=1,M. \tag{2.5}$$

Dynamic equations of motion for these coefficients can be obtained by differentiating (2.5) with respect to the time. Assuming that the order of differentiations may be interchanged, we obtain

$$\dot{Z}_{i,k} = \frac{d}{dt}\left(\frac{\partial c_i}{\partial p_k}\right) = \frac{\partial}{\partial p_k}(\dot{c}_i) = \frac{\partial R_i}{\partial p_k} + \sum_{j=1}^{N} \frac{\partial R_i}{\partial c_j} Z_{j,k} ,$$

$$i,j=1,N, \qquad k=1,M. \tag{3.3}$$

Considering the solutions c_i as elements of a column vector of length N, we can think of the arrays $Z_{i,k}$, $\dot{Z}_{i,k}$, and $R'_{i,k} \equiv \partial R_i/\partial p_k$ either as matrices of N by M elements <u>or</u> as sets of M column vectors each of length N. The Jacobian matrix $J_{i,j} \equiv \partial R_i/\partial c_j$ is a square matrix of N by N elements. If we view the set of N by M equations as M vector equations, we can write

$$\left\{\dot{Z}\right\}_k = \left\{R'\right\}_k + J \cdot \left\{Z\right\}_k , \qquad k = 1,M, \tag{3.4}$$

where it is evident that the columns of $Z_{i,k}$ are completely uncoupled from each other.

Important features of this type of sensitivity analysis application are:

- Eqs. (3.3) can be automatically compiled and solved for arbitrary chemical mechanisms in the same manner as the chemical kinetics equations (3.2)[7]. This eliminates tedious and error-prone hand-programming and thus allows very rapid turnaround for solving arbitrary chemical mechanisms, in both the kinetics and the sensitivity programs.

- Although the M vector equations (3.4) are decoupled from one another, both R' and J depend explicitly on the current values of c_i which are determined by solving (3.2). To obtain fully reliable solutions, each of the M equations (3.4) should be solved simultaneously with (3.2), resulting in a system of 2N simultaneous differential equations to be solved to obtain each of the M column vectors making up $\dot{Z}_{i,k}$.

- The cost of simultaneous solutions, using modern stiff ODE solvers,[8] for the combined set of kinetics and sensitivity equations greatly exceeds the cost of solving the kinetics system separately from the sensitivity equation system. It appears possible to reliably accomplish this separation in many photochemical problems, although some additional research is still needed on this aspect of the problem.

- The method is first order. Although equations can can be similarly derived for higher order sensitivity coefficients, $Z^{(2)}_{i,jk}$, $Z^{(3)}_{i,jk\ell}$, etc., no consensus has yet emerged on the value which would be derived from higher order evaluations. (For many research users, first order terms provide sufficient information, whereas for some policy applications the contributions of higher-order terms may also be desirable. (See Eq.(2.4).)

- Sensitivity coefficients are evaluated continuously over the entire time domain of interest.
- When statistically significant information about the distribution of input parameters is available, output variances, valid to first-order, can be evaluated.[9]

(ii) Chemical Kinetics with Time-Varying Rate Coefficients

When rate coefficients are varying in time (e.g., diurnally varying photochemical rates) the sensitivity coefficient is defined as

$$Z = (\partial/\partial\varepsilon)\left[c_i(c_1,\ldots,c_N;\ p_k(t)+\varepsilon g_k(t);t\right]\Big|_{\varepsilon=0}, \quad (3.5)$$

where the function $g_k(t)$ measures the uncertainty in $p_k(t)$. The sensitivity equations are then given in analogy to Eq. (3.4), by

$$\dot{Z} = R(t) + J\cdot Z. \quad (3.6)$$

The functional derivatives $R_k(t)$ can be evaluated, for the kth parameter, in variational calculus, by

$$R_k(t) = (\partial/\partial\varepsilon)\left[R(c_1,\ldots,c_N;\ p_k(t) + \varepsilon g_k(t);\ t\right]\Big|_{\varepsilon=0} \quad (3.7)$$

This method has been applied for diurnal variations in a simplified atmospheric ozone example by Dickinson and Gelinas[7], and specific choices for the function $g_k(t)$ have been indicated. The important features of the diurnally varying examples were qualitatively much the same as in constant rate coefficient case which was discussed above, but with some operational distinctions.

- The evaluation of functional derivatives requires some dedicated programming of the functions $g_k(t)$ in the sensitivity program.

- Kinetics and sensitivity solutions can be much more costly to obtain when rate coefficients are varying with time than when alternative constant rate coefficients are used. For example, experience in diurnal phothchemistry has shown[10] that diurnal kinetics integrations can be ten to fifty times more costly than corresponding constant (daily averaged) rate photochemistry. This proves to be a serious practical constraint for all dedicated and non-dedicated sensitivity methods.

(iii) Non-Reactive Hydrodynamics

Examples of variational sensitivity methods, as well as the Fourier Amplitude Sensitivity Test, which is a sampling method, have recently been presented and compared by Koda *et al*[11]; they present several computational examples for the atmospheric diffusion of inert species while also discussing reactive systems. The governing non-reactive hydrodynamics equations are (in one spatial dimension).

$$\frac{\partial c_i(\underline{x},t)}{\partial t} = \frac{\partial}{\partial x}\left[K_D(x)\,\frac{\partial c_i(x,t)}{\partial x}\right], \qquad i = 1,N, \tag{3.7}$$

where $K_D(x)$ represents spatially varying diffusion coefficients, which are assumed to be identical for all species. Boundary conditions are represented by

$$-K_D(0)\,\frac{\partial c_i}{\partial x} = S_i(t) \qquad \text{at } \underline{x} = 0, \tag{3.8}$$

where S_i represents all sources and sinks of the i-th chemical species at time t at the boundary $\underline{x} = 0$, and

$$\frac{\partial c_i}{\partial x} = 0 \qquad \text{at } \underline{x} = H(t), \tag{3.9}$$

where H(t) is a time varying boundary height. Initial conditions, $c_{i,I}(x) \equiv c_i(\underline{x}, 0)$, are usually assumed to be known with no error.

Sensitivity equations are obtained by first discretizing Eq. (3.7) in the spatial domain, such as by the method of lines.[12] The partial differential equations (3.7) are thereby reduced to a large, finite set of ordinary differential equations in which $c_i(x,t)$ and $K_D(x)$ are evaluated only at discrete spatial locations x_ℓ, $\ell = 1,L$. For a single species the discretized form of Eq. (3.7) is (in vector notation, and including boundary conditions)

$$\frac{dC(t)}{dt} = A(K_D)\cdot C(t) + S(t), \qquad (3.10)$$

where $C(t) = \left[C(x_1,t),\ C(x_2,t)\ \ldots .C(x_L,t)\ \right]^T$, $K_D = \left[\ K_D\ (x_1),\ K_D(x_2),\ldots K_D(x_L)\right]^T$, and $S(t) = \left[S(x_1,t),0,..,0\right]^T$. Initial conditions are denoted by $C_o = C(0)$, and the matrix $A(K_D)$ is a finite difference representation of the diffusion terms in Eq. (3.7). For constant K_D, the analytic solution for Eq. (3.10) is

$$C(t) = e^{A(K_D)t}\left[C_o + \int_o^t e^{-A(K_D)t'}\ S(t')dt'\right] . \qquad (3.11)$$

Sensitivity coefficients for this case are defined by

$$Z_j(t) = \left[\frac{\partial C(x_1,t\ )}{\partial K_{Dj}}\ ,\ \frac{\partial C(x_2,t)}{\partial K_{Dj}}\ ,\ldots .,\ \frac{\partial C(x_L,t)}{\partial K_{Dj}}\right]^T , \quad j=1,L. \qquad (3.12)$$

where $K_{Dj} \equiv K_D(x_j)$. A sensitivity equation (in vector form) is obtained by a derivation analogous to that used in the development of Eqs. (3.3) and (3.4). This development yields the sensitivity equations

$$\frac{dZ_j(t)}{dt} = A(K_D)\cdot Z_j(t) + \frac{\partial A(K_D)}{\partial K_{Dj}} . C(t),\ j = 1,L, \qquad (3.13)$$

which can also be solved analytically.

Important features of this method are similar to those mentioned in (i) above, namely:

- The solutions to Eq. (3.10) and (3.13) can be evaluated either analytically or numerically for arbitrary applications.
- The diffusion coefficient at each spatial grid point is treated as an individual, uncertain parameter.
- The method is first order. Higher order terms have not yet been evaluated for this case, to the best of our knowledge. (The utility of higher-order terms is application-dependent.)
- Sensitivity coefficients are evaluated continuously in time over the entire space and time domain of interest.
- This method can be generalized to also include (constant) wind advection terms. In such cases, the wind values assigned to each grid point are also treated as individual, uncertain parameters.
- Output variances, valid to first order, can be evaluated when statistically significant information about the distribution of diffusion data is available.

(iv) Reactive Hydrodynamics with Constant Rate Coefficients and/or Diffusion Coefficients

Sensitivity coefficients are defined for the i-th species (in vector form) by

$$Z_{i,j} \equiv \frac{\partial}{\partial p_j} C_i(t) = \frac{\partial}{\partial p_j} \left[C_i(x_1,t),\ldots,C_i(x_L,t)\right]^T,$$

$$i=1,N, \quad j=1,(M+L), \qquad (3.14)$$

and the spatially discretized sensitivity equations are given by:

$$\frac{\partial Z_{i,j}(t)}{\partial t} = \left[\frac{\partial A(K_D)}{\partial p_j} \cdot C_i + \frac{\partial R_i}{\partial p_j}\right] + \left[A(K_D) + J\right] Z_{i,j}(t),$$

$$i=1,L, \quad j=1,(M+L). \qquad (3.15)$$

In this case, p_j spans all chemical rates $k_1 \ldots k_M$ and diffusion coefficients $K_{D,1}, \ldots, K_{D,L}$ at all spatial grid points. The total number of parameters is (M + L).

To our knowledge, applications at this level of complexity have not yet been done for air quality simulation models. The important features follow the same general lines as in the cases which have previously been discussed. Clearly, the amount of dedicated programming and data management can approach prodigious proportions for this level of applications where there can easily be 30 chemical species, 75 photochemical reactions, and hundreds of grid points.

(v) Reactive Hydrodynamics with Space and Time Varying Coefficients

Cases in which $K_D(\underline{x})$, $S_i(t)$, $H(t)$, and k_j can vary have been considered by Koda *et al*[11]. Equations have been developed for the functional derivatives, $\delta c_i(\underline{x}',t')/\delta K_D(x)$. $\delta c_i(\underline{x}',t')/\delta S_j(t)$, $\delta c_i(\underline{x}',t')/\delta k_j$, and $\delta c_i(\underline{x}',t')/\delta H(t)$. Perturbations about nominal values $K_D^o(\underline{x})$, $S_i^o(t)$, k_j^o, and $H^o(t)$ are introduced which lead to deviations δc_i about a nominal value c_i^o. In the one-dimensional case, first order variational equations (obtained by decomposition of $c_i = c_i^o + \delta c_i$) for the case in which $H(t) = H =$ constant are

$$\frac{\partial}{\partial t}\,\delta c_i(\underline{x},t) = \frac{\partial}{\partial x}\left[K_D^o(\underline{x})\,\frac{\partial\,\delta c_i(\underline{x},t)}{\partial x} + \delta K_D(\underline{x})\,\frac{\partial c_i^o(\underline{x},t)}{\partial x}\right] + \sum_{j=1}^{N}\frac{\partial R_i^o}{\partial c_j}\,\delta c_j(\underline{x},t) + \sum_{j=1}^{M}\frac{\partial R_i}{\partial k_j}\,\delta k_j, \quad i=1,N. \tag{3.16}$$

subject to initial conditions

$$\delta c_i(\underline{x},0) = 0, \qquad (3.17)$$

and boundary conditions

$$-K_D^o(0)\frac{\partial \delta c_i(\underline{x},t)}{\partial x} - \delta K_D(0)\frac{\partial c_i^o(\underline{x},t)}{\partial x} = \delta S_i(t) \text{ at } \underline{x} = 0, \qquad (3.18)$$

and

$$\frac{\partial \delta c_i(\underline{x},t)}{\partial x} = 0 \qquad \text{at } \underline{x} = H, \ i=1,N. \qquad (3.19)$$

Porter[13] suggested that input parameters be expressed as expansions of orthogonal basis functions, which leads to the following development of sensitivity equations:

(a) Select a set of functions $\{\psi_i(\underline{x},t)\}$.

(b) Multiply Eq. (3.16) by ψ_i and sum over i = 1,N.

(c) Integrate the resultant equation in (b) over the intervals $\underline{x} = [0,H]$ and $t = [0,T]$.

(d) Apply the initial and boundary conditions, Eqs. (3.17) - (3.19).

(e) Specify a defining equation for $\psi_i(\underline{x},t)$

$$\frac{\partial \psi_i}{\partial t} = -\frac{\partial}{\partial x}\left(K_D^o(\underline{x})\frac{\partial \psi_i}{\partial x}\right) - \sum_{j=1}^{N}\frac{\partial R_j^o}{\partial c_i}\psi_j, \qquad i = 1,N, \qquad (3.20)$$

with

$$\frac{\partial \psi_i}{\partial x} = 0, \ \underline{x} = [0,H]. \qquad (3.21)$$

(f) Specify a terminal condition on the $\{\psi_i\}$, e.g.,

$$\psi_i(\underline{x},T) = \delta_{i,j}\delta(\underline{x} - \underline{x}'), \quad i,j=1,N, \tag{3.22}$$

where δ_{ij} and $\delta(\underline{x} - \underline{x}')$ are, respectively, the Kronecker and Dirac delta functions.

The results of performing operations (b) and (c) and solving Eq. (3.20) gives

$$\delta c_i(\underline{x}',T) = -\sum_{j=1}^{N} \int_0^H \delta K_D(\underline{x}) \int_0^T \frac{\partial c_j^o}{\partial x} \frac{\partial \psi_{ji}}{\partial x}\, dt\, dx$$

$$+ \sum_{j=1}^{N} \int_0^T \delta S_j(t)\, \psi_{ji}(0,t)\, dt$$

$$+ \sum_{j=1}^{M} \delta k_j \sum_{\ell=1}^{N} \int_0^H \int_0^T \frac{\partial R_i^o}{\partial k_j} \psi_{\ell i}\, dt\, dx. \tag{3.23}$$

The functional derivatives which we seek are

$$\frac{\delta c_i(\underline{x}',T)}{\delta K_D(\underline{x})} = \sum_{j=1}^{N} \int_0^T \frac{\partial c_j^o}{\partial x} \frac{\partial \psi_{ji}}{\partial x}\, dt\,, \tag{3.24}$$

$$\frac{\delta c_i(\underline{x}',T)}{\delta S_j(t)} = \psi_{ji}(0,t)\,, \tag{3.25}$$

$$\frac{\delta c_i(\underline{x}',T)}{\delta k_j} = \sum_{\ell=1}^{N} \int_0^H \int_0^T \frac{\partial R_\ell^o}{\partial k_j} \psi_{\ell i}\, dt\, dx\,. \tag{3.26}$$

A similar exercise can be carried out in order to obtain $\delta c_i(\underline{x}',t')/\delta H(t)$ when H is a function of time. Sensitivity functions (3.24) to (3.26) are obtained by solving a decomposed equation for the nominal values $c_i^0(\underline{x},t)$, and solving Eq. (3.20) for $\psi_{\ell i}(\underline{x},t)$ in order to carry out the operations in (3.24)-(3.26). Koda *et al* [11] have obtained solutions for

$$\frac{\delta c(\underline{x}'t)}{\delta K_D(\underline{x})}, \quad \frac{\delta c(\underline{x}',T)}{\delta S(t)}, \text{ and } \frac{\delta c(\underline{x}',T)}{\delta H(t)}$$

for a single non-reacting species in a 1-D (vertical) boundary layer simulation and have related the resulting functional derivatives at specific points to sensitivity functions obtained in a discretized grid calculation which was discussed in (iii) above. The relationship at spatial location $\underline{x}'$ between sensitivity coefficients which are developed from functional derivatives and those which are developed from discretized grid point formulations is

$$Z_{ij}(\underline{x}',t) \equiv \frac{\partial c_i(\underline{x}',t)}{\partial p_j(x_\ell)} \cong \int_{x_\ell - \frac{1}{2}\Delta x}^{x_\ell + \frac{1}{2}\Delta x} \frac{\delta c_i(\underline{x}'t)}{\delta p_j(x)}\, dx, \qquad \begin{matrix} i=1,N, \\ j=1,L, \end{matrix} \quad (3.27)$$

$$\frac{\partial c_i(\underline{x}',t)}{\partial p_j(x_\ell)} \cong \Delta x \frac{\delta c_i(\underline{x}',t)}{\delta p_j(x_\ell)}, \quad (3.28)$$

where Δx represents an appropriate grid spacing and $\delta p_j(x)$ is assumed to be zero at grid points other than x_ℓ.

The major features to note for the variational methods involving functional derivatives are:

- The solution of the system equations for nominal values, $c_i^o(\underline{x},t)$, and of the adjoint equations for $\psi_{\ell i}(\underline{x},t)$ are complex calculations, even with no reactions present; but the method produces highly rigorous sensitivity measures.
- This highly dedicated method will be difficult to automate sufficiently for reliable, rapid turnaround calculations in arbitrary model applications.
- The method is first order and subject to the same considerations which were previously discussed. No higher order evaluations have yet been made.
- Sensitivity coefficients are evaluated at specific space and time points $(\underline{x}',t')$ but rigorously contain correlative effects which have occurred along characteristic trajectories which lead to $(\underline{x}',t')$.
- Arbitrary deviations are the measure of sensitivity in this method. It is not yet apparent that expectation values of observables could be readily obtained when meaningful parameter statistics are available.

The Green's Function Method

Hwang, Dougherty, Rabitz, and Rabitz[14] have developed a variant of the direct variational method of sensitivity analysis for chemical kinetics systems. The basic development of the method is similar to that described in our discussion of the variational method for chemical kinetics with constant rate coefficients.

Starting from the kinetics model equations (3.2), the dynamic equations for the evolution of the sensitivity

coefficients $Z_{i,k}$ are derived as before, obtaining equations (3.3) or (3.4). As we remarked in the earlier discussion, Eq. (3.4) can be viewed as M uncoupled equations, each describing the evolution of a column vector of length N:

$$(\dot{Z})_k = (R')_k + J\cdot(Z)_k \ , \qquad k = 1,M. \qquad (3.4)$$

In the most general applications of the direct method, each one of these equations is solved numerically together with the basic kinetics equations (3.2) in order to allow the stiff equations system solvers to adjust the time steps optimally for the numerical integration of each of the vector equations in (3.4), without introducing numerical errors due to interpolation from an independent solution run for the basic kinetics equations (3.2).

The Green's function method continues its development by assuming that each of the vector equations in (3.4) can be reliably and safely solved by itself, interpolating values of c_i obtained by a careful separate integration of (3.2). Since Eq. (3.4) is linear in Z, the solution of the inhomogeneous equation can be expressed in terms of a Green's function $K_{i\ell}(t,\tau)$ as

$$Z_{i,k}(t) = K_{i,k}(t,0) + \int_0^t \sum_{\ell=1}^{N} K_{i\ell}(t,\tau)\ R'_{\ell k}\ (c_1,\ldots c_N;\ \{p\};\tau\ )\, d\tau,$$

$$i,\ell=1,N,\ \ k=1,M, \qquad (3.29a)$$

if p_k is one of the initial conditions, where $R'_{\ell k} \equiv \partial R_\ell / \partial p_k$; or as

$$Z_{i,k}(t) = \int_0^t \sum_{R=1}^{N} K_{i\ell}(t,\tau)\ R'_{\ell k}\ (c_1,\ldots,c_N;\ \{p\};\tau)\ d\tau \qquad (3.29b)$$

if p_k is any other parameter of the system. In either case,

the Green's function is the solution of the homogeneous equation

$$\dot{K}_{ik}(t,\tau) - \sum_{\ell=1}^{N} J_{i\ell}\,(c_1,\ldots,c_N;\{p\};t)\,K_{\ell k}(t,\tau) = 0,$$
$$i,\ell,k = 1,N, \qquad (3.30)$$

where $J_{i\ell} \equiv \partial R_i/\partial c_\ell$ is the Jacobian matrix as before, subject to the causality condition

$$\underline{K}\,(t,\tau) = 0 \quad \text{for all } t<\tau, \qquad (3.31)$$

and the initial value condition

$$K_{ik}(\tau,\tau) = \delta_{ik}, \qquad (3.32)$$

where δ_{ik} is the Kronecker delta.

Note that (3.30) may be treated as a set of N decoupled equations for N column vectors of length N. Provided the numerical errors introduced by interpolating values of $c_i(t)$ obtained by solving (3.2) are acceptable, significant computational savings can be achieved by solving (3.30), an N x N system, N-times, instead of solving either a 2N x 2N system (for coupled direct methods) M-times, or an N x N system (for decoupled direct methods) M-times, for any system which has more uncertain parameters than it has chemical species (M>N), which is usually the case for realistic simulation models.

Practical implementation of the Green's function method proceeds as follows:

(a) The simulation model equations (3.2) are integrated numerically using the nominal values $\{p^o\}$ for the uncertain parameters for times t=0 to t=T, recording or storing values for the solutions c_i at numerous intermediate points in time.

(b) The time interval T is subdivided by a coarse grid of I+1 quadrature times τ_m, $\tau_o \equiv 0 < \tau_1 < \tau_2 < \ldots < \tau_I \equiv T$, which are selected with due consideration for details of the solutions $c_i(t)$.

(c) Eq. (3.30) is solved numerically for each of the subintervals $\tau_{m-1} \leq t \leq \tau_m$ with initial conditions $K_{ik}(\tau_{m-1}, \tau_{m-1}) = \delta_{ik}$. The values of $c_i(t)$ needed to evaluate $J_{i\ell}(t)$ are interpolated as needed from the stored solutions.

(d) The sensitivity coefficients themselves are computed by using the appropriate form of Eq. (3.29), replacing the integrations over time by numerical quadratures on the coarse grid τ_m. It can be shown that the Green's function for any two points on the mesh may be obtained as a product of the Green's functions obtained for the intervening subintervals:

$$K_{i,n}(\tau_q, \tau_r) = \sum_{j=1}^{N} \cdots \sum_{m=1}^{N} K_{i,j}(\tau_q, \tau_{q-1})\, K_{j,k}(\tau_{q-1}, \tau_{q-2}) \cdots \cdots K_{\ell,m}(\tau_{r+2}, \tau_{r+1}) K_{m,n}(\tau_{r+1}, \tau_r) \qquad (3.33)$$

(e) If higher order sensitivity coefficients are desired as well, they may also be computed by quadratures with the same Green's function:

$$Z_{i,jk\ldots n}(t) = \int_0^t d\tau \sum_{\ell=1}^{N} K_{i\ell}(t,\tau) \left\{ D_j D_k \cdots D_n R_\ell(\{c\},\{p\},\tau) - J_{i\ell}(\tau) Z_{\ell,jk\ldots n}(\tau) \right\}, \quad i=1,N,\ j,k,\ldots,n=1,M; \qquad (3.34a)$$

where the differential operator D_j is defined as

$$D_j \equiv \frac{\partial}{\partial p_j} + \sum_{q=1}^{N} Z_{q,j} \frac{\partial}{\partial c_q} \quad . \qquad (3.34b)$$

The method is clearly a dedicated method, requiring new programming for each problem, but it is highly amenable to automatic programming methods just as the chemical kinetics equations are.

SAMPLING METHODS

Monte Carlo Method

Stolarski *et al*[15] have recently presented an atmospheric uncertainty analysis which is based upon Monte Carlo selection of trial values for chemical rates in a 1-D stratospheric model which gives steady state solutions of Eqs. (2.1) with $\underline{v} = 0$. Many of the photochemical reactions of stratospheric interest have been measured often enough and well enough that it is reasonable to assign probability distributions for the parameters describing these reactions. Most rates which are known to be significant in the stratosphere are uncertain within factors ranging from 1.1 to 10.

Hypothetical reactions must simply be included and assigned nominal values and uncertainties on a tentative basis. The Monte Carlo method is executed as follows (see Figure 5):

(a) A log-normal distribution is assigned to the probability that true reaction rates lie between (k_j^o/r_j) and $r_j k_j^o$, where $r_j > 1$ is the factor by which k_j is uncertain.

(b) The atmospheric model is run, using a chosen "best set" of reaction rate values and center line values for other parameters.

(c) A random number generator is used to select trial values for the entire set of reaction rates, simultaneously.

(d) The atmospheric model is run again, using the set of simultaneously perturbed trial reaction rates from (c).

(e) The processes (c) and (d) are repeated until enough output samples are accumulated to evaluate statistically meaningful mean values and variances of the output variables. Stolarski *et al*(15) ran sufficient cases so that the accumulated 1σ imprecisions on both sides of the output distributions converged to within 1 or 2%.

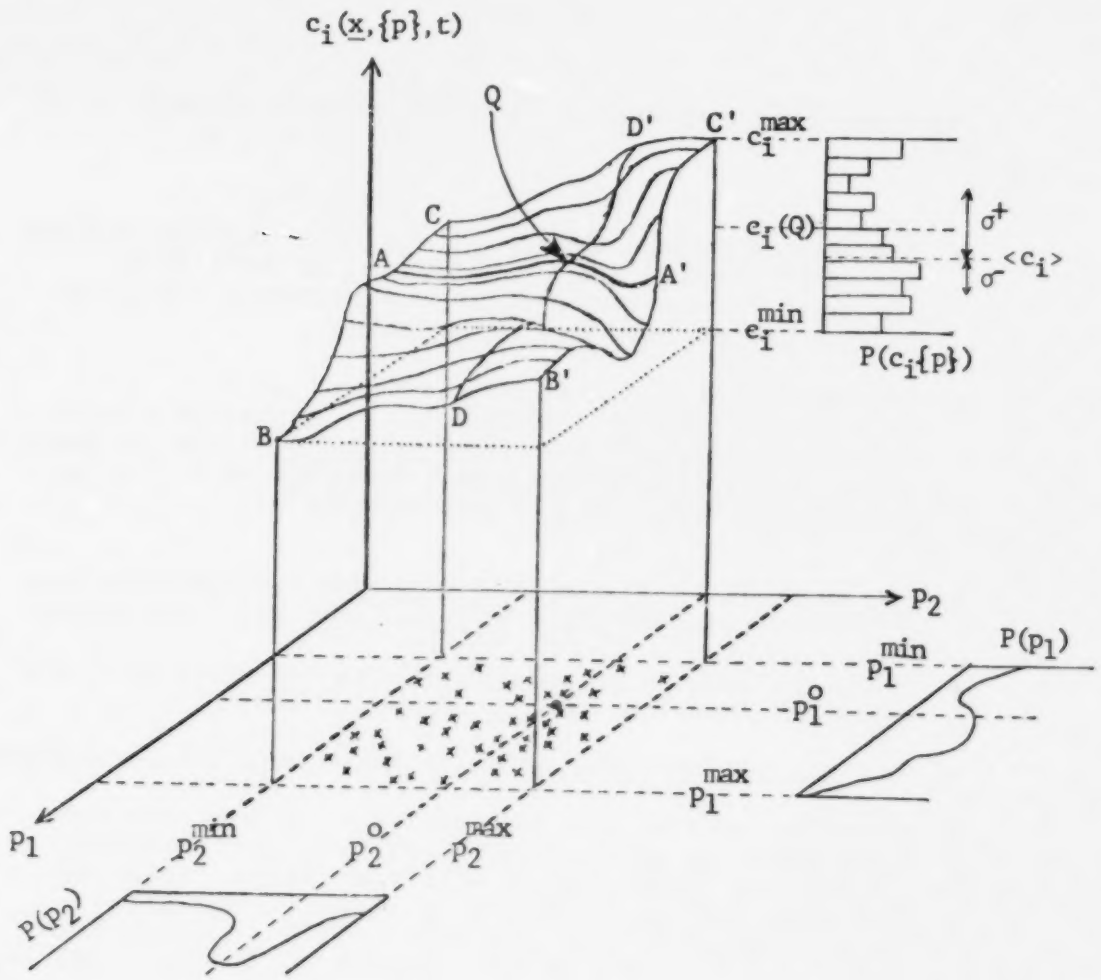

Figure 5. Schematic illustration of the Monte Carlo method of sensitivity analysis. A random number generator is used to select values of all the uncertain parameters within the domain of uncertainty. The simulation model is then run using those values for the parameters. Values thus computed for $c_i(\underline{x},\{p\},t)$ are then tallied and analyzed by standard statistical methods. The distribution of values obtained in this way is illustrated above as a histogram, with mean value $\langle c_i \rangle$ and standard deviations σ^+ and σ^-. Stolarski *et al* assume log-normal distributions for the uncertain parameters, but any distribution function could be used, including a joint probability distribution for two or more correlated parameters.

The important features of the Monte Carlo method are:

- The method is non-dedicated and thus readily applies to arbitrary air quality models. To date only chemical rate uncertainties and boundary condition uncertainties have been considered.
- The method has, so far, been applied as an uncertainty analysis method which evaluates output variances. Questions of how a shift in one parameter value would change the model response to all other parameters have not yet been indicated.
- Dependences of the method upon selected probability distribution functions have not yet been indicated.
- Best and worst case estimates can be identified and retained from the accumulated test cases.
- Initial studies are underway for correlated parameters.
- Computer economics presently require that the 1-D stratospheric model be physically simplified and solved for steady-state solutions rather than the far more costly transient solutions. Stolarski *et al* emphasize that the feasibility of using the Monte Carlo method depends most heavily upon reducing the computational cost of the atmospheric model to an absolute minimum. In their case, complete reprogramming and physical simplifications were required for their original, more general atmospheric model; this went far beyond simply altering the original model and obtaining nominally improved economies.
- Sensitivities were determined separately by the time-honored method applied to individual rates. The rates were varied, one at a time, by $\pm$ 1σ and by $\pm 2\sigma$.
- The number of test cases which were required to obtain statistical convergence in $\sigma_i(x_\ell, t)$ was proportional, in some sense, to the number of sensitive parameters rather than to the total number of uncertain parameters. Convergence was more rapid for those species and locations which were most affected by rates with small imprecision; relatively poor convergence was experienced for species such as the methane oxidation chain, for which important reaction rates are quite uncertain.

- Stolarski *et al*[15] emphasized that the uncertainty analysis must be redetermined when: (i) a new reaction is included in the model (ii) substantial changes occur in parameter values or their imprecisions, or (iii) substantial changes occur in the basic understanding of atmospheric processes.

FOURIER AMPLITUDE SENSITIVITY TEST METHOD

The Fourier Amplitude Sensitivity Test was first developed by Cukier and Shuler *et al* [16] to provide a means for determining the sensitivity of the solutions $c_i(\underline{x}, \{p\}, t)$ of a large set of coupled rate equations to uncertainties in each parameter p when averaged (in a certain sense) over uncertainties in all the other parameters.

Figure 6(a) illustrates the basic idea of the FAST method. For simplicity in illustration, consider a system of coupled rate equations depending on only two parameters, p_1 and p_2. Although the investigator can select some "best value" for each of these parameters, the true value lies within some range of uncertainty about that "best value".

If only one parameter were uncertain, we could profitably ask, "What is the slope of the solution surface as that single parameter is varied over its range of uncertainty?" (See line AA' in Figure 6(a).) But other parameters are also uncertain; thus in the general case we do not know where on the solution surface we are. It is thus of some value to ask, "What is the average slope of the solution surface in the direction of parameter p_j when we average over the full range of uncertainties in all other parameters as well?" In other words, what is the average slope of curves AA', BB', CC', etc.?*

---

*In some cases, the average slope may be zero or very small for parameters which in fact are sensitive. The average slope transverse to a symmetrical trough or ridge, for example, vanishes, even if the trough is very deep (or the ridge very high). To avoid being mislead by such cases, a squared norm is sometimes used to indicate averaged sensitivities.

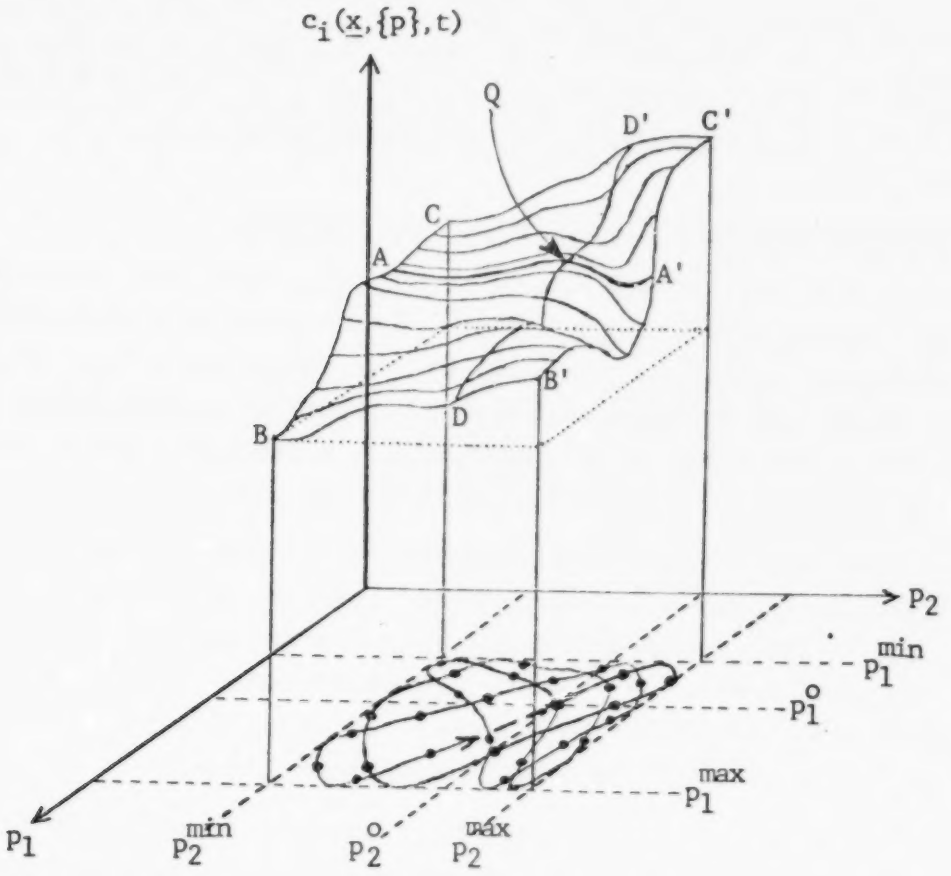

Figure 6(a). Schematic illustration of the Fourier Amplitude Sensitivity Test (FAST). The FAST algorithm generates a closed search curve across the domain of uncertainty of the parameters which begins at the "best values" of all parameters. Values of c_i at each sampling point (marked by large dots in the figure) are tabulated against the search parameter s along the search curve to produce a periodic function of s as shown in Figure 6(b). Note that the sampled points shown here do not include the extreme values of c_i (points B and C').

In principle, one could determine the slopes of the solution surface along each of the parameter axes at a large number of points on the solution surface above the domain of uncertainty of the parameters and compute suitable

averages. Such brute force methods, indeed, have been used on occasion for systems involving only a few parameters and small numbers of coupled differential equations. The FAST algorithm, on the other hand, provides a technique for selecting much smaller numbers of sampling points. The method is most likely to be successful for those systems in which the solution surface is smooth and preferably monotonic, with minimal structure in the way of deep valleys, sharp ridges, or peaks close to potholes.

The method consists of four steps. First, a set of M integer frequencies ω_ℓ is selected subject to certain constraints to be discussed below. Second, one frequency is assigned (arbitrarily) to each of the parameters considered to be uncertain, setting

$$p_\ell = p_\ell^o \exp \{u_\ell(\sin \omega_\ell s)\} \qquad (3.35)$$

where s is a parameter of variation; p_ℓ^o is the "best value" of p_ℓ; and u_ℓ is a continuous function of its argument selected so that its extreme values result in values for p_ℓ at the extremes of the range of uncertainty for p_ℓ. Note that p_ℓ is then periodic in the parameter s; as s varies, the point $\underline{p}$ determined by (3.35) moves about in the domain of uncertainty of all the parameters. If the set of frequencies ω_ℓ are mutually prime, the point $\underline{p}$ traces out (exactly once) a closed curve (a Lissajous figure) within the domain of uncertainty as s ranges from 0 to 2π.

Third, the values of p_ℓ resulting from setting $s = 2\pi q/N^*$; $q = 1, 2, 3, \ldots, N^*$ are used in running the air quality simulation model from given initial conditions to time t. The values of the solutions $c_i(\underline{x},\{p(s)\},t)$ are stored after each simulation run. N^* is proportional to the largest frequency ω_ℓ.

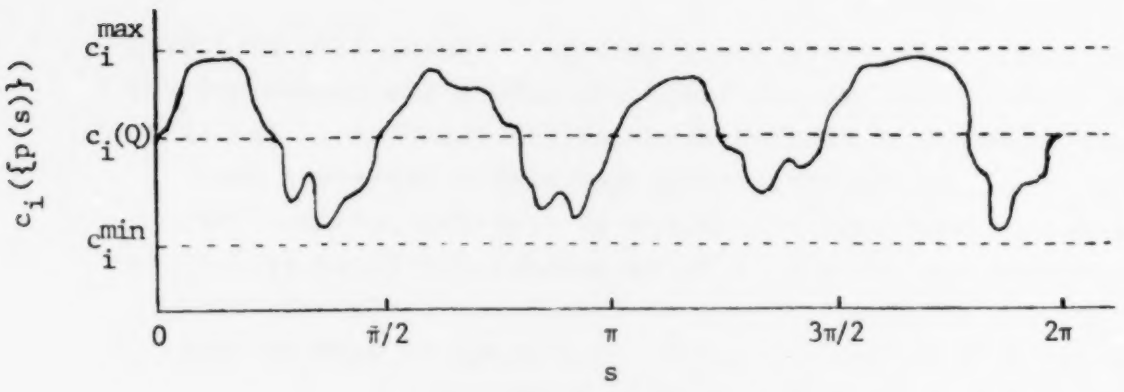

Figure 6(b). The solution $c_i(\{p(s)\})$ is a periodic function of the search parameter s. The Fourier amplitudes of this function at the frequencies assigned to any parameter p_i provide an estimate of the average slope of the solution surface in the direction of the p_i-axis. The search curve does not in general reach the extreme values of c_i.

Fourth, the solutions c_i for a given position and time $(\underline{x},t)$ are now periodic functions of s itself (Figure 6(b).) Fourier analysis of this function yields amplitudes corresponding to each of the integer frequencies ω_ℓ :

$$A^i_{\omega_\ell} = \frac{1}{2\pi}\int_0^{2\pi} c_i(\{\mathbf{p}(s)\})\sin(\omega_\ell s)\,ds \quad . \tag{3.36}$$

Only sine components occur in the expansion of $c_i(p(s))$ because of the symmetries of Eq. (3.35). This integral can be approximated by a simple summation over the accumulated solutions:

$$A^i_{\omega_\ell} \approx \frac{2}{N^*}\sum_{q=1}^{N^*} c_i(\{p(s_q)\})\sin(\omega_\ell\, s_q) \quad . \tag{3.37}$$

The Fourier amplitude $A^i_{\omega_\ell}$ measures the contribution made by variations in p_ℓ to the total variations in c_i as all the parameters p oscillate back and forth across the domain of uncertainty.

It is thus not surprising that the Fourier amplitudes defined above can be shown to be proportional to a weighted average of the partial derivatives of c_i with respect to each parameter:

$$A^i_{\omega_\ell} \propto \int_{p_1^{min}}^{p_1^{max}} \int_{p_2^{min}}^{p_2^{max}} \cdots \int_{p_M^{min}}^{p_M^{max}} P(\underline{p}) \frac{\partial c_i}{\partial p_\ell} \, dp_1 \, dp_2 \cdots dp_M$$

$$\propto \langle Z_{i,\ell} \rangle \quad , \tag{3.38}$$

the averaging being performed over the volume of the domain of uncertainty of the parameters.

In Eq. (3.38) above, $P(\underline{p})$ measures the distribution of sampling points $s = 2\pi q/N^*$ across the domain of uncertainty. For convenience in computing the summation (3.37), the sampling is done at equal intervals of the parameter s. But the functions $u_\ell(s)$ are highly arbitrary. They are required to be continuous and single-valued, but no other constraints are necessary except that the extreme values of u_ℓ results in the extreme values of the parameter's domain of uncertainty, and that $u_\ell = 0$ results in the "best value" p_ℓ^o of that parameter. As indicated in Figure 7, however, a wide variety of functions $u_\ell(s)$ can satisfy these minimal constraints. Selecting sampling points at equal intervals in s will result in sampling distributions over the uncertainty ranges of the parameters p whose shape depends explicitly on the assumed shape of $u_\ell(s)$; the resulting Fourier amplitudes will also vary depending on the investigator's choices for $u_\ell(s)$ and on the regularity of the solution surface $c_i(\underline{x},\{p(s)\},t)$.

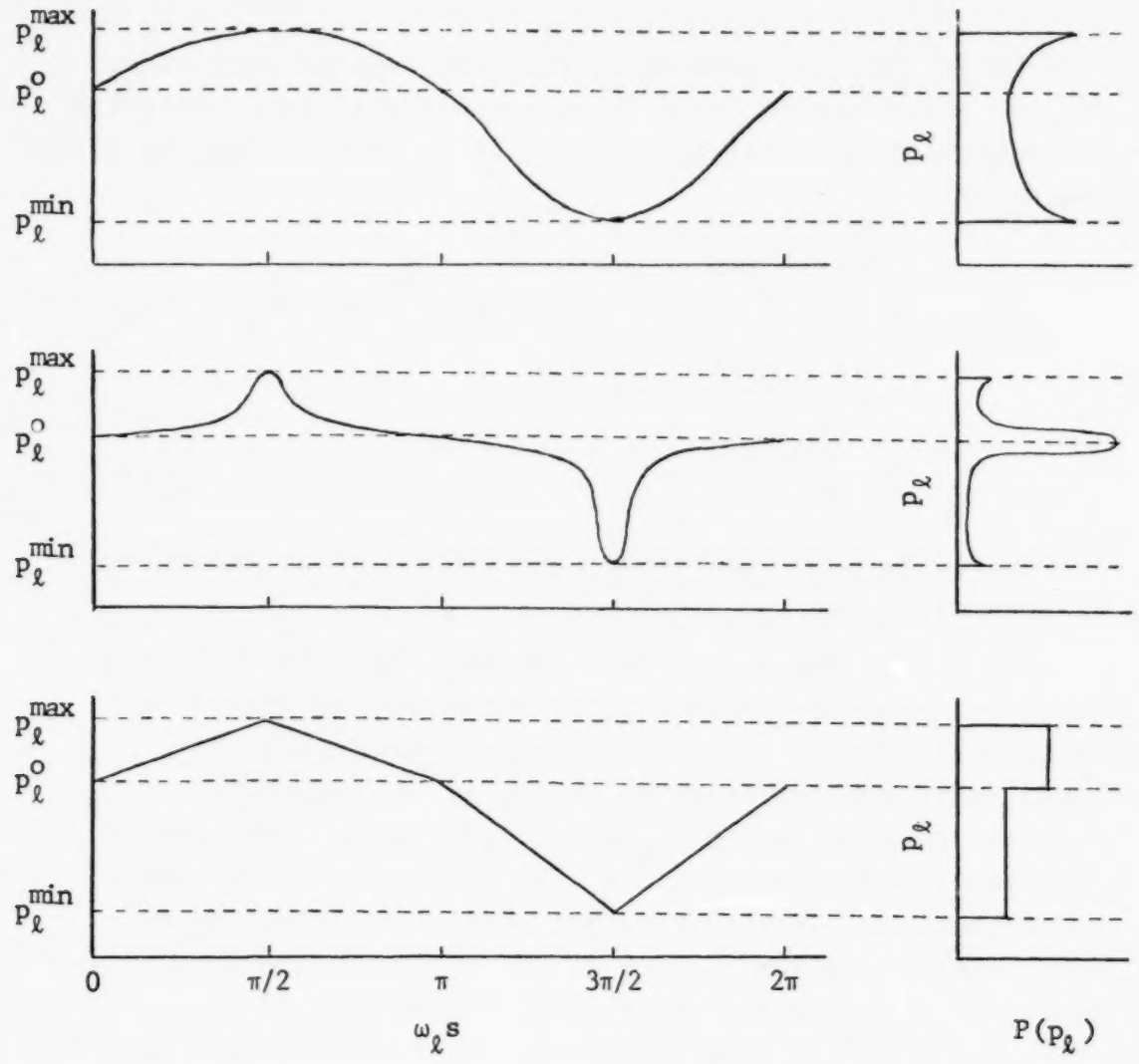

Figure 7. The functional forms used to generate the search path in the FAST algorithm are highly arbitrary, resulting in a great deal of ambiguity about the probability distributions $P(p_\ell)$.

The number N* of simulation runs required is determined by the highest frequency in the set $\{\omega_\ell\}$, since each cycle of a sinusoidal wave must be sampled at several points in order to determine the amplitude of that Fourier component. For economy, we would like N* to be as small as possible; for accuracy in evaluating the amplitudes, we would like N* to be as large as possible. If we make N* larger, then we can also increase ω_{max}, the largest frequency in the set, which has the effect of folding the search curve back and forth in the domain of uncertainty more times, increasing the likelihood of detecting fine structures in the solution surface and of the search curve coming very close to the highest and lowest points (the extremes) of the solution surface.

Various algorithms have been developed for generating sets of frequencies for use with FAST. The frequencies should be chosen to minimize ambiguities in the determination of Fourier amplitudes which arise from interferences between frequencies or harmonics. As a minimum, the frequencies must be linearly independent:

$$\sum_{i=1}^{M} a_i \omega_i \neq 0 \qquad (3.39)$$

for integer coefficients a_i subject to

$$\sum_{i=1}^{M} |a_i| \leq M_I + 1, \qquad (3.40)$$

where M_I is an integer. Such a set of frequencies will then be free of interferences to order M_I.

Because the Fourier amplitudes are computed from a finite number N* of equally spaced sampling points, "aliasing" can also confuse the picture. Aliasing occurs whenever a sum of two or more frequencies (and/or their harmonics) is equal to an integer multiple of the number of sampling points:

$$\sum_{i=1}^{M} a_i' \omega_i = JN^* \qquad (3.41)$$

where the coefficients a_i' and J are also integers. To minimize such aliasing, we require

$$\sum_{i=1}^{M} a_i' \omega_i \neq JN^* \qquad (3.42)$$

with

$$\sum_{i=1}^{M} |a_i'| \leq M_I+1. \qquad (3.43)$$

The frequency set $\{\omega_\ell\}$ will then be free of aliasing to order M_I. For $M_I = 4$, it has been found empirically that the maximum frequency and the number N* of sampling points necessary are proportional to $M^{2.5}$. No analytic expression has yet been obtained for N* in terms of M and M_I; such a formula would have to be derived by the techniques of combinatorial algebra which would have to provide first of all an explicit algorithm for generating the set of frequencies $\{\omega_\ell\}$.

The important features of the FAST method are:

- It is a non-dedicated method which requires no alterations of the air quality models.
- It provides relative measures of expected sensitivities. To date, only first-order measures, $\langle Z_{i,k} \rangle$, have been obtained in applications, although higher order information is potentially available should there be a need or interest in abstracting it.
- Applications to date have emphasized pure kinetics or pure non-reactive hydrodynamics.
- Consideration of correlated parameters is in the early stages.
- Computer economy of individual air quality simulation models is a major factor in whether or not a sufficient number of cases can be accumulated as adequate input to the FAST method.

- Variances obtained by summing all specific frequency components are generally inexact since only a finite number of terms can ever be computed numerically.
- The method can ultimately serve the interests of both policy-makers and of scientific researchers.
- Each application of the FAST method provides sensitivities at a specific time point, in snapshot fashion, rather than continuously in time as do the deterministic methods. (The same is true for other sampling methods, as we saw for the Monte Carlo method.)
- In difficult problem regimes, the results obtained depend upon specific sampling distributions, as is evident from considering Figure 7 and as observed in practice by Penner.[19] If the solution surfaces are very irregular, different search patterns yield different output results.
- Given a sufficiently dense search pattern, the method can in principle resolve subtle system sensitivities associated with complex wrinkles or pathological features in the solution surface. (The extent to which this possibility is ever realized in practice is problematical due to considerations of computer economics as discussed in Section 4.)

TABLE 1

SUMMARY TABLE

CHARACTERISTICS OF CANDIDATE SYSTEMATIC SENSITIVITY AND UNCERTAINTY ANALYSIS METHODS

METHOD	VARIABLES	PROGRAMMING FEATURES	APPLICATIONS TO DATE	REQUIRED INPUT AND EXECUTION FOR M PARAMETERS	COMMENTS
DETERMINISTIC METHODS (A.) TIME-HORIZONED	$\Delta c_i(\underline{x},t)$; $Z_{i,j}$, $Z^{(2)}_{i,jk}$	Non-Dedicated-Results are obtained as continuous functions of time for all variables.	Models from essentially all scientific and engineering disciplines--pure kinetics, 1-D, 2-D, and 3-D transient models.	Minimum of (M+1) air quality model integrations-includes both nominal and perturbed parameter values. All parameters can be tested.	This is the favored method of most practical users. No special programs are required. The trial-and-error concept is intuitively appealing and logically transparent to a majority of users. No statistical considerations are required. All model parameters, whether constant or varying can be tested. Results are amenable to best and worst case identification.
(B.) VARIATIONAL	$Z_{i,j}$, Δc_i(first order)	Dedicated-Results are obtained as continuous functions of time for all variables. Sensitivity model should be decoupled from air quality model.	Only pure kinetics and 1-D models. No higher dimensional systems.		Coming into widespread use as automated programs become applicable to arbitrary problem scenarios. Requires no pre-screening of parameters for atmospheric kinetics. No statistical considerations are required, and $Z_{i,j}$ and Δc_i (first order) are conceptually transparent. Appears to be more economical than all other alternative methods (in both human effort and computer costs). Results are amenable to best and worst case identification.
• Constant parameters (direct method)					
(i) pure kinetics		(i) Highly automated	(i) atmospheric chemistry, chemical lasers, and combustion chemistry	One integration of the air quality model using nominal parameter values plus M integrations of sensitivity equations.	
(ii) hydrodynamics (reactive and non-reactive)		(ii) only 1-D transient sensitivity models have been solved with minimal automation for arbitrary problems. 2-D transient sensitivity models are now reasonably near at hand with highly automated features due to recent advances in FEM methods.	(ii) 1-D atmospheric dynamics. Further developments can be readily made in 1-D, whereas somewhat more extensive developmental efforts are still required for 2-D applications.		
• variable parameters (functional derivatives)				The above integrations plus solutions of adjoint sensitivity equations are are required, plus some double integrals must be computed. In addition to being more costly, these operations are difficult to automate for arbitrary problem scenarios.	Functional derivatives require significant hands-on control by the user, and are considerably more costly to solve than cases with constant parameters. In practical applications it would be most reasonable to approximate time variations by piecewise constant variabilities so that a more efficient direct method can be used in place of functional derivatives.
(i) pure kinetics		(i) Semi-automated	(i) diurnal photochemistry for the simple Chapman mechanism.		
(ii) pure hydrodynamics		(ii) Not-automated	(ii) Test problems for a diffusive boundary layer.		
SAMPLING METHODS					
(A) Monte Carlo	$\sigma_i(\underline{x},t)$	Non-Dedicated. Results are obtained at single time points for all variables	1-D steady state stratospheric ozone model	On the order of $(\gamma)^{M_s}$ model integrations, where γ is the number of sample points per parameter and M_s is the number of sensitive parameters. Generally $\gamma \geq 2$ and $M_s < 20$ for computer economy. All model parameters can be tested, but applications to date emphasize chemical rates.	Results are expectation values which are generally dependent upon assumed parameter distributions. Pre-screening is necessary to estimate in advance the number of sensitive parameters which in turn govern the rate of convergence of the method ($\sigma \propto (\gamma)^{M_s}$). Results are good to all orders and are conceptually transparent. Results are not amenable to best and worst case identification unless raw sample data are additionally processed. Even with pre-screening estimates, the number of samples required to give convergence is not known exactly for each specific $\sigma_i(\underline{x},t)$; monitoring is required during evaluation of σ_i's.
(B) FAST	partial variances and $\langle Z_{i,j} \rangle$	Non-Dedicated. Results are obtained at single time points for all variables.	Models from several scientific disciplines and perhaps socio-economic models. Purely kinetics and 1-D non-reactive diffusion are the major atmospheric applications to date.	On the order of $(M)^{2.5}$ model integrations are required using sample values of perturbed parameters. All model parameters can be tested given suitable sampling (distribution) functions.	Results are expectation values which are generally dependent upon assumed parameter distributions. Prescreening may be necessary to limit the number of tested parameters to practical limits (say 20 or fewer parameters for pure kinetics and fewer still for more complex models with higher execution costs). Sampling method is generally more efficient than the search method used in the Monte Carlo method. Can handle both constant and time varying parameters. Results are not amenable to best and worst case identification unless raw sample data are additionally processed.

SECTION 4

EVALUATION OF CANDIDATE SENSITIVITY METHODS FOR AIR QUALITY MODEL APPLICATIONS

This section illustrates "what you get" and "what you pay" with systematic sensitivity and uncertainty analysis methods in their present states of development. Evaluations of the candidate systematic sensitivity and uncertainty analysis methods are given in the discussion below in terms of our detailed assessments of the performance characteristics of each method for: (i) chemical models, (ii) 1-D transient models and (iii) 3-D air quality models. The results are self-evident and indicate a mixed picture--- one in which both reasonable and unreasonable options are avaiable. Accordingly, several criteria are used for our evaluations:

- Motivations. Fortunately, "what you get" can be very much a function of what you want or need in choosing systematic sensitivity and uncertainty analysis methods. Optional methods are available for those with policy-making motives and for those in atmospheric research. These respective motivations will largely determine the selection of candidate analysis methods for air quality simulation models.
- Cost. This can be the make-or-break factor, involving not only computer execution costs but also human programming costs. Human programming costs include the development of new sensitivity programs (for dedicated methods) as well as the essential tasks of preparing input data and writing preprocessor and graphics routines. For large, complex regional air quality models the human costs alone can prove to be prodigious depending upon specific applications, as can computer execution expenses. This is true

both for dedicated sensitivity programs, per se, and for the multitudes of air quality model runs which may be needed as input to non-dedicated sensitivity methods. For smaller, purely photochemical kinetics box models or for purely hydrodynamics models, computer costs are more reasonable as are human programming costs when automated compilation features are used.

- Usefulness. We particularly focus on criteria which underlie whether or not potential users may sooner or later be motivated to use candidate systematic sensitivity and uncertainty analysis methods. In addition to labor and computer economy, it is essential that a method be conceptually transparent and that its results be readily interpretable. (This factor alone has accounted for a majority of practical users continuing to stand by the time-honored method. The overwhelming appeal of the time-honored method derives from several factors; it is highly intuitive in nature, it requires no special preparations to operate initially, and no special operations are needed to interpret output results.) An additional criterion of utility is the potency of candidate analysis methods. Here, the major promise of some systematic sensitivity methods is their ability to identify more effectively and to evaluate quantitatively subtle sensitivities in complex solution surfaces. Potent methods also have the capacity to anticipate unexpected events (new discoveries or other surprises.)

  Whether or not potency of a method should be associated with the availability of higher-order sensitivity measures is a subject of continuing debate. On the one hand, policy makers desire reliable variances (uncertainty measures) for the results of air quality models. The

most reliable means of obtaining such information is by sampling results from repetitive runs of the air quality model (basically, the time-honored or Monte Carlo method). It is clearly out of the range of practicality to construct variances by first evaluating and then summing terms involving $Z_{i,j}$, $Z^{(2)}_{i,jk}$, etc., from either the deterministic or the FAST methods. On the other hand, users who require sensitivity measures are generally quite satisfied with first-order sensitivity coefficients (regardless of the methodology which is used to generate the coefficients) because first-order coefficients contain all of the information which most users want and need on a value-for-value basis in practical applications.*

With some reflection, all of these criteria are recognized to be branches of the same tree, the tree of user acceptance. This should not be surprising because these issues of user acceptance have perpetually governed the rate of development and the pattern of use of systematic sensitivity and uncertainty analysis methods. Tables 2 and 3 provide a summary of our evaluations of the candidate analysis methods. Detailed discussions of these evaluations follows directly below.

BASIC CASES

Three basic examples are used for purposes of illustrating evaluations in this section. In all cases, computer cost

---

* This view has been expressed repeatedly by the overwhelming majority of scientific practitioners; it represents, in fact, the hard reality of the situation. This does not say that higher-order sensitivities are insignificant--we have clearly shown the contrary in previous sections--but rather that, on a value-for-value basis, first-order sensitivity coefficients provide a major part of the information which practical users seek from sensitivity analysis.

estimates assume that a Control Data Corporation (CDC) 7600 computer is used for sensitivity and uncertainty computations at a cost of $1200 per execution hour ($20/minute). This is obviously an arbitrary basis, but it can be converted approximately to the appropriate, equivalent basis for other computer systems. The main points of our evaluations should be quite evident within this selected context despite the great variability of computer execution times for the various models and computers which are used by different investigators.

A. Chemical Models (purely kinetics)

This base case assumes that:

- Thermal and photochemical rates are constant. (Time-varying rates are far more costly to integrate using existing ODE system solvers)
- External chemical sources are constant.
- The system has N chemical species and M reaction rate parameters.
- Numerical integration of the ODE system of kinetic (species) equations has costs which are proportional to N^{α}. It is reasonable to assume that $\alpha \approx 2\text{-}3$, which is representative of implicit solution methods for stiff ODE systems, such as variants of the Gear method[8]. Generally, a system of 20 species would require approximately 15 seconds of CDC 7600 execution time at a cost of about $5 per kinetics run; a system of 50 species requires about 4 minutes CDC 7600 execution time at an approxinate cost of $80 per kinetics run. Kinetics execution times are not very sensitive to the number of reaction rate parameters, M.
- Automatic kinetics programs are available which allow users to select, compile, and solve arbitrary reaction sets (with arbitrary rates), requiring virtually no dedicated programming.[17]

B. 1-D Transient Atmospheric Models

One-dimensional transient models have been applied primarily to reactive plumes and to global stratospheric ozone modeling. In addition, 1-D models provide a convenient foundation for subsequent considerations of 2-D and 3-D regional air quality simulation models. This base case assumes that:

- thermal and photochemical rates are constant over intervals of one hour or longer. (Instantaneous variations greatly increase execution costs).
- External chemical sources are constant over intervals of one hour or longer.
- The system has N chemical species, M parameters, and L spatial zones.
- The cost of numerical integrations depends upon the choice of solution methods. (For example, simultaneous space-time solutions using conventional matrix methods are far more costly than space-and-time split solutions using Gauss elimination methods.) Current models tend to require on the order of several minutes of CDC 7600 execution time per simulation when simultaneous space-time solution methods are used.
- Automatic compilations of arbitrary chemical mechanisms, velocities, and diffusion profiles are possible, but are not yet in widespread use.
- N = 20, L = 50, M = 150. (We assume there are 50 chemical rates, 50 velocity values, and 50 diffusion coefficient values at the respective grid points, resulting in M = 150.) Initial and boundary conditions will be considered in later examples.

C. 3-D Regional Air Quality Models

This base case assumes that:

- Thermal and photochemical rates are constant over intervals of one hour or longer
- The spatial grid is 25 x 25 x 5, for a total of 3125 elements.
- N = 10
- The system has 25 chemical rates, of which 5 are photodissociation.
- 3 velocity components (measured at 3 hour intervals)
- Diffusion is characterized by horizontal and vertical components K_H and K_V.
- The model treats 5 emission factors (NO_x, CO, 3 hydrocarbons), measured hourly.
- Initial values are available for all species, the temperature, and the relative humidity (R.H.).
- Boundary values are available at all surfaces.
- While numerical integration costs are highly dependent upon choices of solution methods, current models of this type (which split space and time integrations) require approximately 30 minutes of CDC 7600 execution time for a 24 hour simulation.

CHEMICAL MODEL EVALUATION

A. The Time-Honored Method

(i) Sensitivities

What can be simpler than merely altering the value of any parameter p_j, while holding the remaining parameters at their nominal values, and directly noting the resulting variations in all solutions, $\delta c_i(t)$, for $i=1,..N$, continuously in time? This is the essence of the time-

honored method. The performance characteristics of this method are:

- Statistical properties of parameter distributions are not required.
- Values of $\delta c_i(t)$ are accurate to all orders.
- Values of $\delta c_i(t)$ can be obtained with equal facility for time-varying rate coefficients as for constant rates. (The cost of diurnal kinetics runs can exceed constant parameter runs by factors of 5 to 50.)
- A minimum of (M + 1) kinetics runs are required--one run for the nominal parameter set and one run for each rate which is varied. (Assume all M are to be varied.) In practice, several trial values may be required in order to locate the sensitive range of some of the parameters, which frequently leads to a cost of $\sim$ 2M kinetics runs. For 20 species and 50 rate parameters, the cost of 2M runs would be approximately \$5 x 2 x 50 = \$500. For 50 species and 100 rate parameters, the corresponding cost would be \$80 x 2 x 100 =\$16,000 (for testing all 100 parameters).
- Potentially new discoveries or unknown rates can be tested by including hypothetical reactions in the basic mechanism and then determining values of their rates which would be required in order for the hypothetical reaction to become significant in the overall mechanism.
- Whenever new nominal rate values are used or new reactions are included in a mechanism, the sensitivity analysis must be repeated *in toto*.

- Sensitivity coefficients can be obtained as

$$Z_{i,j} = \lim_{\Delta p_j \to 0} \frac{\Delta c_i}{\Delta p_j} ,$$

$$Z^{(2)}_{i,jk} = \lim_{\Delta p_j, \Delta p_k \to 0} \frac{\Delta^2 c_i}{\Delta p_j \Delta p_k} , \text{ etc.}$$

  Recall, however, that it is very poor economy to evaluate sensitivity coefficients as a means of obtaining δc_i (see Eq. 2.3) to high-order accuracy. It is far more effective to get Δc_i directly--by simply introducting any desired perturbation Δp_j and using the time-honored method.

- Expectations of sensitivity coefficients, $\langle Z_{i,k} \rangle$, can be obtained by accumulating a huge number of outputs from kinetics runs in which all permutations of simultaneous rate perturb.tions have been introduced. This requires on the order of $[\gamma]^M$ kinctics runs where γ is the number of trial perturbations to be applied to each parameter. If only two trial values are used for each of 50 parameters, $[2]^{50}$ kinetics runs would be required, which is clearly out of the range of practicality.

(ii) Uncertainties

- Variances σ_i can be evaluated from data accumulated in the same fashion as was mentioned immediately above. In this case Stolarski et al[15] have found that the number of samples is much less than $[\gamma]^M$. This will be discussed further below under Monte Carlo Methods.

- Best case and worst case estimates for δc_i can be evaluated by directly applying selected combinations of parameter perturbations. The choices of parameter perturbations are arbitrary, and the process of finding best and worst case parameter combinations is not unique; exhaustive trials appear to be required.

B. Variational (Direct) Methods

These methods are most appropriate for evaluating sensitivity coefficients $Z_{i,j}$. The performance characteristics for first-order sensitivity computations are:

- The sensitivity equations (3.3) can be automatically compiled and solved for arbitrary chemical mechanisms. The solutions can be obtained either simultaneously with the chemical kinetics equations or decoupled from them. Decoupling must be carefully performed and thoroughly validated against the simultaneous case; this is the subject of current research.

- The costs for decoupled sensitivities are about the same as a kinetics run, which goes as N^3. This amounts to (MN^3) for a system of M parameters plus one kinetics run at nominal parameter values which gives a total cost proportional to $(M+1)N^3$. For 20 species and 50 parameters, this implies a cost of approximately \$250; for 50 species and 100 parameters, the approximate cost is \$8000. The costs of solving the sensitivities and kinetics simultaneously is proportional to $M(2N)^3$ for all its parameters, or about 8 times as expensive as the corresponding decoupled case. The approximate costs of the simultaneous case are \$2000 for 20

species and 50 parameters, and $64,000 for 50 species and 100 parameters. Clearly the redundant kinetics solutions which characterize the simultaneous case should be avoided.

- Costs of evaluating systems with time-varying coefficients are inordinately greater, both in machine execution time and in dedicated programming of functional derivatives.
- Whenever new nominal values or new reaction sets are used, the sensitivity analysis must be reperformed.
- Expectations of sensitivity coefficients $\langle Z_{i,j} \rangle$ are not practical to evaluate. Estimates of δc_i and σ_i based on first-order sensitivity coefficients have only local validity (see Section 1.)

C. Green's Function Method

This method is similar in many respects to the variational (direct) methods just discussed, but has not been tested or applied as extensively as yet. Key performance characteristics of this method are as follows:

- The homogeneous differential equations (3.30) for the Green's function are easier to solve (in terms of computation time) than are the sensitivity equations (3.13) of the direct method, and can be automatically compiled and solved for arbitrary chemical mechanisms.
- The numerical solution of (3.30) must be reinitialized repeatedly, adding some expense to the computation. Additional expense is incurred in computing the sensitivities $Z_{i,k}(t)$ from the Green's function by quadrature (trapezoidal rule or equivalent). These added costs, however, should be smaller than the savings achieved by decoupling the sensitivity equations from the kinetics equations.

- Whether or not the decoupling itself can be done without losing accuracy remains to be proven for reasonably complicated kinetics systems.
- If desired, higher order sensitivity coefficients can be obtained at far less cost than by the direct method.
- Additional programming efforts totalling perhaps one person-year should allow the development of a user-ready package which could resolve these questions.

D. The Monte Carlo Method

This is the extension of the time-honored method which computes variances of model output data. Performance characteristics are as follows:

- This method is conceptually transparent; it is non-dedicated; and it applies to one time point at a time.
- $[\gamma]^M$ kinetics runs are required as input, where γ is the number of trial perturbations applied to each parameter. For $\gamma=2$, the following dollar costs would be incurred for (constant parameter) kinetics runs alone:

20 Species

50 parameters - $\$ 5.6 \times 10^{15}$
100 parameters - $\$ 6.3 \times 10^{30}$ $\}$ $\simeq$ infinite

50 Species

50 parameters - $\$ 9 \times 10^{16}$
100 parameters - $\$ 1 \times 10^{32}$ $\}$ $\simeq$ infinite

For all practical purposes, these costs are infinite, suggesting that more selective parameter sampling

must be used. One alternative would be the FAST sampling scheme. So far as we can determine, this possibility has not yet been explored in a practical example. We will see in (E) directly below that even the FAST sampling would be inordinately expensive in kinetics integration for our basic examples. Stolarski *et al*[15] estimate in advance the likely order-of-magnitude cost of variance evaluations. They apply the time-honored method, using trial perturbations of $\pm\sigma$ and $\pm 2\sigma$ for all rates, in order to make a preliminary identification of the most sensitive parameters in the system. (Recall that this is a local evaluation, varying one rate at a time about nominal parameter values, which is good to arbitrary order and is continuous in time.) In the event that the kinetics mechanism is sensitive, say, to only 10 rates out of the original set of 50 or 100 rates, then only $(2)^{10}$ or 1024 kinetics integrations are required as input for $\gamma=2$. In this event the cost of the input for computing variances would be $5120 for 20-species systems and $81,920 for 50-species systems. This approach is now a naturally-evolved hybrid of sensitivity and uncertainty analysis. This practical alternative will prove useful for all of the systematic methods. Whereas in the Monte Carlo method, prescreening only serves to estimate execution costs, in the FAST method, prescreening followed by rejection of insensitive parameters may result in considerable savings of input samples without compromising the basic results. This point should be further pursued and either validated or invalidated by FAST users.

- In cases where prescreening is used for parameter rejection, the hybrid approach exacts a cost elsewhere,

namely, some of the promise which systematic methods hold for a more rigorous search of model solution surfaces is compromised by computer economics.

- The entire analysis must be redone if any nominal values change. In some cases, changes in the magnitude of parameter uncertainties (probability distributions) may also require that the entire analysis be redone.
- Sensitivities tested with perturbations of $\pm\sigma$ do not sample the "corners" or extreme regions of the solution surfaces. Best case and worst case examples are more likely to escape notice in these circumstances.
- Stolarski *et al* found that output variances were essentially independent of the insensitive parameters. But the rate of convergence to reliable variances was dependent upon the degree of sensitivity of particular variables in various time (and space) domains. Hence the user of the Monte Carlo method can not know *a priori* the number of input runs which are required to obtain convergence of variance values.

E. Fourier Amplitude Sensitivity Test

The performance characteristics for first-order sensitivity coefficients are:

- The FAST is a non-dedicated method which can be applied to arbitrary model outputs at one time point of the model.

- The number of kinetics integrations which are required as input to the FAST is approximately $(M)^{2.5}$. Therefore, approximately 17,700 kinetics runs are required for 50 parameters; and 100,000

kinetics runs are required for 100 parameters. The following dollar costs would be incurred for (constant parameter) kinetics runs alone:

20 Species

50 parameters - $ 88,500

100 parameters - $ 500,000

50 Species

50 parameters - $1.42 x 10^6

100 parameters - $8. x 10^6

- It is clearly necessary to minimize the number of parameters under consideration as well as to minimize the cost of reliably solving kinetics systems.

- A conundrum arises. The promise of systematic methods is to search the model solution surfaces more thoroughly in order to detect subtle as well as obvious reaction coupling influences. Increasingly thorough searching, in turn, suggests that more and more complete reaction mechanisms should be put to the test. But the computer economies argue strongly in the opposite direction, thereby thwarting some of the promise of systematic methods which happen to be expensive.

- Variances can be obtained by two means:

  (i) The output of the numerous kinetics runs which are required by the FAST can be processed directly. It is known that the parameter space is sampled non-uniformly, depending upon the specific choice of FAST distribution functions. The effect of sampling choices upon such output variances has not yet been assessed, but is likely to be significant for sensitive parameters.

(ii) The FAST obtains the sensitivity measures $\langle Z_{i,j} \rangle$ from partial variances $\sigma_i(p_j) \equiv \sigma_i(\omega_j, t)/\sigma_i$. The terms $c_i(\omega_j, t)$ are defined in terms of the Fourier amplitudes at frequency ω_j by

$$\sigma_i(\omega_j, t) = \frac{2}{\sigma_i} \sum_{\alpha=1}^{\infty} \left\{ A^{(i)}_{\alpha\omega_j}(t)^2 + B^{(i)}_{\alpha\omega_j}(t)^2 \right\} . \qquad (4.1)$$

In practice only a finite number of ω_j and associated harmonics are actually used, so that the resulting variance values may or may not be reliable.

- Unaveraged sensitivity measures $Z_{i,j}$ (analogous to measures obtained by the deterministic methods) can be obtained in principle by using distributions in the FAST of zero width (zero uncertainty) for all $p_{k \neq j}$, but this would be very inefficient relative the other sensitivity methods for kinetics systems.
- In principle, second-order and perhaps higher-order sensitivity information is available from the FAST. In practice, this capability does not yet appear to have been implemented for computations by general users. As discussed above, this additional information will not provide a more effective means of obtaining improved variances *vis à vis* other alternatives; and the application of higher-order, averaged sensitivity information appears to be a very low priority need of most practical users, due to problems of conceptual transparency and ease of application. (It is our impression that this is the case for higher-order sensitivity information from all existing methods)

• The sensitivity measures $\langle Z_{i,j} \rangle$ depend in general upon the choices of distribution functions, as was discussed above and also found by Penner[18] in applications. The sampling methods thus tend to be best-suited to problems in which probability distributions are well-known for key parameters.

1-D TIME DEPENDENT MODEL EVALUATION

From the evaluation of chemical models just above, it is clear that computer economics will dominate our evaluations of sensitivity methods for 1-D and higher-dimensional transient air quality models. The cost of integrating multi-dimensional transient models can be highly variable and is particularly dependent upon the numerical methods and programming efficiency in any specific air quality model. To provide a basis for subsequent evaluations we will first discuss briefly a set of well-known numerical solution options which are used in multi-dimensional transient models and then indicate approximate scale relationships of the costs of these solution options to the costs of purely chemical models which were previously stated.

Transient 1-D chemical-transport models are generally solved simultaneously in $(\underline{x},t)$ by employing the method of lines to discretize spatial gradient operators. An implicit stiff ODE integration method is then applied to solve the overall system of size $\left[NL \times NL\right]$. When no special numerical advantages are explcited, the cost of solving such a 1-D model is proportional to $\left[NL\right]^{\alpha}$, where α has a value between 2 and 3. When central differences are applied to gradient operators, resulting in an overall matrix having a block-tridiagonal structure; Gauss elimination solutions of the matrix can be used which lead to reduced solution costs on the order of LN^{α}.

[By similar analysis, simultaneous solutions in ($\underline{x}$,$\underline{y}$,t) in 2-D transient models (e.g., LIRAQ) have solution costs ranging from L^3N^α to $[L^2N]^\alpha$. (For the remainder of this discussion we can assume $\alpha \tilde{\sim}$ 2-3, but the structural aspects are clearer if we scale costs in terms of α.) The use of ADI (alternating direction implicit solution) methods for 2-D transient models reduces the numerical solution cost to the range of $2L^2N^\alpha$ to $2L[LN]^\alpha$. In 3-D air quality models (e.g., the Systems Applications, Inc., regional model), ADI is absolutely essential. Solution costs for 3-D air quality models would then scale to span the range of $3L^3N^\alpha$ to $3L^2[LN]^\alpha$ (for Gauss elimination versus conventional matrix solution options).]

For estimation purposes, we will assume that 1-D models are solved simultaneously in ($\underline{x}$,t) at a cost of 5 minutes of CDC 7600 time, or $100 per simulation. Again, this estimate is representative of existing transient 1-D models with which we are familiar; the main thrust of our conclusions will not be greatly affected by this particular choice of cost estimate *vis a vis* execution costs of other such 1-D transient atmospheric models actually in operation today.

A. The Time-Honored Method

Assume that, in addition to the single model solution which is required using nominal parameter values, only one additional run will be required for each parameter in order to evaluate $\delta c_i(\underline{x},t)$. (This might be the case for a very experienced practitioner.) A minimum number of parameters would be M = 150, which is composed of:

- 50 chemical rates which are constant in time and have the same value in every zone. Assume that the same rate perturbation applies simultaneously to every zone.

- 50 diffusion coefficients which are uncorrelated and take specific values at each zone. Assume that a single fractional perturbation at each zone is appropriate over the entire time history.
- 50 wind velocities which are uncorrelated and take specific values at each zone. Assume that a single fractional perturbation at each zone is appropriate over the entire time history.

In practice, many other parametric variations may also be considered, including

- pollutant emission sources at each zone as a function of time;
- boundary values of species concentrations or species fluxes as a function of time;
- initial conditions at every zone; and
- photodissociation rate variations from zone-to-zone due to variable cloud cover (even for constant rate coefficients).

On this minimum basis, the cost of 1-D air quality model runs alone would be (151 x $100) or $15,100 of computer time. (The reader can readily make similar estimates which are germane to his own computing circumstances and experiences.) Major improvements in computer economy could be realized from possible advances in four areas. First, improved numerical techniques such as matrix solutions by Gaussian elimination would probably bring the computational cost under $1000 for 150 model runs. Second, intuitive prescreening for obviously insignificant sensitivities can reduce the number of parameters to be varied; the utility of this option depends strongly upon the craft of the user. Third, correlated variables can sometimes be identified and varied

simultaneously rather than individually. Fourth, optimized programming, using automatic compilation of arbitrary problem scenarios, machine coding, parallel processing, and other specialized program optimization routines, can result in substantial savings. For example, Stolarski et al[15] found it necessary to write a completely new stream-lined production version of their original 1-D atmospheric model and further, to reduce it to a steady-state model rather than a transient model. However, this latter option would probably not be appropriate for multi-day air quality simulations which are intrinsically transient in nature.

B. Variational Methods

Using the same basic example as above, several operational possibilities can be dismissed immediately. For example, simultaneous solution of the sensitivity and 1-D model equations is out of the question. (Recall the discussion for Chemical Models.) Similarly, the sensitivity equations would be solved for one parameter at a time rather than solving the larger matrix for, say, all M parameters simultaneously. Further, automatic compilation for arbitrary problem scenarios is essential for both the sensitivity programs and 1-D air quality models. (Can you imagine the difficulty of writing a new dedicated sensitivity program which corresponds perfectly to each air quality model and/or to each specific application which is of interest?)

Following these suggestions and assuming that solutions of the sensitivity equations would cost the same to obtain as solutions of the 1-D air quality model, one would be solving a system of size $[NL \times NL]$ (M+1) times. A practical approach to calculating sensitivities for this problem (with constant chemical rates) might be to pursue the following lines:

- For a given set of nominal $\underline{v}^o$ and $\underline{\underline{K}}_D^o$ at each grid point, solve the sensitivity equations M_R times for the reaction rates of interest. This gives $Z_{i,j}|_{chem.}$ at all grid points continuously in time.
- For a given set of nominal chemical rates, solve the sensitivity equations $M_{\underline{v}}$ and $M_{\underline{\underline{K}}_D}$ times for the velocities and diffusion coefficients at selected grid points of interest. ($\underline{v}$ and $\underline{\underline{K}}_D$ may be constant over, say, hourly intervals; this would pose no difficulty to the direct methods.) This procedure gives $Z_{i,j}|_{Vel.}$ and $Z_{i,j}|_{Diff.}$ at all system grid points, continuously in time.
- For a given set of nominal values for chemical rates, winds, and diffusion coefficients, the sensitivities $Z_{i,j}$ can also be determined at all grid points, continuously in time, for selected initial, boundary, and source conditions.

So far as we can determine, no sensitivity analysis of this level of generality has yet been performed with direct methods. (Koda et al[11] have considered much more limited examples, such as non-reactive atmospheric diffusion, which could be solved analytically.) In the event that M=150 in the general case (due to any combination of values for M_R, $M_{\underline{v}}$, $M_{\underline{\underline{K}}_D}$, and other parameters), the computer cost would be identical ($15,100) to the minimum case discussed for the time-honored method immediately above. Notice that the possibility of using intuitive or other judgements to eliminate obviously insensitive parameters from the systematic analysis is tacitly left open, recognizing:

(i) the element of risk in such eliminations, and

(ii) the fact that some of the promise of systematic methods is compromised to computational economy by such eliminations.

The functional derivative approaches for either time-varying rates or non-discretized spatial variables are severely limited by the following factors:

- The methods are too highly dedicated, at present, for multi-dimensional application.
- 1-D model execution costs increase dramatically for diurnally varying rates and continuously varying emissions sources.
- We estimate execution costs for solving the air quality and the adjoint sensitivity equations to be on the order of solving an $[NL \times NL]$ system of ODE's for the air quality equations plus an $[N^2L^2 \times N^2L^2]$ system of ODE's for the adjoint equations. Thus the execution cost would scale approximately as $[N^2L^2]^{2\alpha}$, with a relatively large proportionality factor applying in the case of variable rates, sources, etc. In addition, N double integrals at, say, L values of $\underline{x}$ would have to be evaluated at each time point for each chemical rate. Likewise, N double integrals for each pair ($\underline{x}'$, $\underline{x}$) and each time point would have to be evaluated for sensitivities to winds and diffusion coefficients. We conclude that, *vis à vis* other methods, this approach is far too costly and difficult to apply in air quality modeling for the foreseeable future.

C. The Monte Carlo Method

For 150 parameters, $(2)^{150}$ runs of the 1-D model is, practically, an infinite input cost. To date this method has been used primarily to obtain steady state atmospheric model variances associated with uncertain chemical rates. Even in the extremely unlikely event that only 10 parameters were found (by some other means) to be sensitive, 1024 runs of a 1-D transient model would be on the order of $100,000 in computer execution cost. This is the reason that Stolarski et al[15] emphasize the need to take every

reasonable step to reduce the execution cost of the atmospheric model which generates the basic data for the Monte Carlo method of evaluating variances. In the event that 20 sensitive parameters are present in an air quality model, over a million input samples would be required to evaluate variances, which is out of the question if air quality model execution costs greatly exceed 1/100 cents per run.

D. The FAST Method

The input cost of testing 150 parameters would be $\approx (150)^{2.5} \approx 275{,}500$ runs of the 1-D air quality model, costing on the order of $27.5 million for computer execution. The non-dedicated features of this method remain very attractive, but the number of parameter trials must be markedly reduced either by pre-screening eliminations or by considering only segments of the overall problem (such as testing the chemical mechanisms independent of the fluid dynamics or vice versa).

3-D AIR QUALITY MODEL EVALUATION

The general picture is abundantly clear by now. If we consider only a 24-hour simulation with the 3-D model at a cost of approximately $600 per run, the computer costs relative to the 1-D examples present a grim picture, indeed. To carry through this discussion, Tables 2 and 3 have presented what we have termed an "Oversimplified Case" and a "Realistic Case" for the number of parameters which must be considered in our basic 3-D air quality model. The "Oversimplified Case" is simplified beyond scientific usefulness. The "Realistic Case" is scientifically acceptable but is well out of the range of practicality. The results in Tables 2 and 3 are self-explanatory.

TABLE 2

SUMMARY TABLE
BASE CASES FOR ILLUSTRATION OF EXECUTION COSTS
OF CANDIDATE SYSTEMATIC SENSITIVITY ANALYSIS METHODS

BASE CASE	DESCRIPTION	ASSUMED EXECUTION COSTS (on basis of "typical" CDC 7600)	COMMENTS
PURE CHEMICAL KINETICS MODELS	(A.) 20 species, 50 parameters (B.) 50 species, 100 parameters	$5/kinetics simulation $80/kinetics simulation	The parameters are assumed to be constant. Diurnal and other time dependent effects would be treated by using piecewise constant parameters over intervals of an hour or longer.
1-D TRANSIENT MODELS	20 species, 50 grid points. (Hence there are 50 diffusion coefficient values and 50 wind values on the basis of one value per grid point and uncorrelated $\underline{v}(\underline{x})$ and $\underline{K}_D(\underline{x})$.)	$100/1-D simulation	Parameters are assumed at least piecewise constant (as above).
3-D REGIONAL AIR QUALITY MODELS	10 species, 3125 grid points (based on zoning which is dimensioned as 25 x 25 x 5), 25 chemical rates, 3 velocity components, $K_{Horiz.}$ and $K_{Vert.}$, 5 emission factors (NO_x, CO, three reactive hydrocarbons, measured hourly), Initial values for species, temperature, and relative humidity. Boundary values at all surfaces.	$600/3-D simulation	

A. Over-simplified Case. 137 parameters, assuming that:

- $\underline{v}$ has identical values at all grid points.
- K_H, K_V have identical values at all grid points.
- No cloud corrections; thus J's do not vary from one grid point to another.
- k's do not vary from one grid point to another (constant T, R.H. at all grid points)
- Initial conditions for 10 species, T, and R.H. are identical at all grid points.
- Boundary conditions of $\underline{v}$, K_H, K_V, and 10 species are identical over each surface. (15 components x 6 surfaces = 90 values)

B. Realistic Case. $\approx$ 234,200 parameters, assuming that:

- $\underline{v}$ is variable at each grid point.
- K_H and K_V are variable at each grid point.
- 5 J's, which are variable at each grid point.
- 20 k's, which are variable at each grid point.
- Initial conditions are variable at each grid point.
- Boundary conditions are variable at each grid point.

TABLE 3

SUMMARY TABLE

ILLUSTRATIVE COMPUTER EXECUTION COSTS FOR CANDIDATE SYSTEMATIC SENSITIVITY ANALYSIS METHODS (REFER TO BASE CASES IN TABLE 3.1(a))

METHOD	PURE CHEMICAL KINETICS MODELS	1-D TRANSIENT MODELS	3-D REGIONAL AIR QUALITY MODELS
TIME-HONORED ($\delta c_i(t)$)	(A.) $500 (Assumes two trials per parameter) (B.) $16,000	$15,100 (assume only one trial per parameter)	(A.) $82,200 (assume one trial / parameter) (B.) $1.4x10^8 (13 yrs CDC 7600 @ 24/hrs/day execution)
DIRECT VARIATIONAL ($Z_{i,j}(t)$)	(A.) $250 (Assumes sensitivity eqs. are decoupled from kinetics eqs.) (B.) $8000 (A.) $2000 (Assumes sensitivity and kinetics eqs. are coupled) (B.) $64,000	$15,100 (assume that sensitivity and 1-D models are decoupled)	(A.) $ 82,200 Plus: unknown number of person-years of dedicated programming effort. (B.) $1.4 x 10^8
FAST ($\langle Z_{i,j}(t')\rangle$)	(A.) $88,500 (Assumes no prescreening) (B.) $8.10^6	$\gtrsim$$27.5 x 10^6 (2.6 years CDC 7600 execution @ 24 hrs/day)	(A.) $132 x 10^6 (12.5 yrs CDC 7600 @ 24 hrs/day) (B.) practically infinite
MONTE CARLO ($\sigma_i(t')$)	(A.) $5.6 x 10^{15} (B.) $1.x10^{32} (these figures are, practically, infinite) (Assumes two samples per rate and all rates are equally sensitive	practically infinite	practically infinite

SECTION 5

TODAY'S APPROACH

The central objective in this section is to suggest the means by which the EPA, today, could most effectively apply systematic sensitivity analysis methods to arbitrary air quality models, which we will assume to be transient 2-D or 3-D regional or reactive plume models. Our suggested choices of method will be guided to a significant extent by the conclusions which have evolved from our evaluations in the previous sections.

CONCLUSIONS

A. On balance, the full promise of systematic sensitivity analysis methods cannot yet be realized in applications to entire 2-D and 3-D transient air quality models. However, by segmenting the models or by prescreening for sensitive parameters, very useful sensitivity information can be gained without significant alterations being required in the air quality models.

B. A combination of time-honored and systematic sensitivity analysis methods can be effectively used on individual air quality model segments; for example just the chemistry can be analyzed in arbitrary zones with the fluid dynamics remaining decoupled, and conversely the chemistry can be frozen while analyzing the fluid dynamics, the boundary conditions, the initial conditions, sources, etc. (Without question prescreening and segmenting compromise some of the promise of systematic methods in exchange for computational economy; this compromise is necessary until further advances occur in systematic sensitivity analysis and/or in the efficiency of execution of air quality models.)

C. Reliable variances appear to be out of practical range for multidimensional transient models, with a multitude of sensitive parameters.

D. Dedicated sensitivity methods must be programmed so that arbitrary problem scenarios can be automatically compiled in a hands-off manner; otherwise, non-dedicated methods are a far more practical alternative for analysis of complex air quality models.

E. For better or for worse, practical users favor conceptually transparent sensitivity methods over those which require more subtle interpretation without providing commensurately more useable information. (For example, higher-order sensitivity coefficients hold important topological information about complex solution surfaces; but the volume of information which is needed to accurately map complex surfaces grows quickly to unmanageable and conceptually obscure proportions.)

F. Numerical efficiency of air quality models and sensitivity methods assumes ever-increasing importance when extensive sensitivity studies on the models are desired. (Most air quality models would require considerable reprogramming to reach their maximum attainable efficiencies for production purposes.)

G. Depending on specific circumstances, field or laboratory measurements may be a preferred alternative to sensitivity analyses. At one extreme are the chemical models where sensitivity analysis is very cost-effective for indicating the most critical reactions which need to be measured in the laboratory in order to markedly improve a chemical mechanism. At the other extreme are 3-D regional models where the costs for even a <u>single</u> sensitivity analysis

may easily exceed $100,000 for computer expense, alone. But we have seen that, in addition, many such analyses must be done to account for alternative parameter choices which are furthermore subject to many permutations. In this case, comprehensive uncertainty analysis of the 3-D model is unreasonable and should be supplanted by more meaningful experimental work. Between these extremes, however, more subtle tradeoffs must be very carefully considered on an individual case basis.

TODAY'S PRACTICAL APPLICATION OF SYSTEMATIC SENSITIVITY ANALYSIS

Variances of air quality model outputs can be economically and reliably obtained for only very special circumstances (such as models with less than about 10 sensitive variables and for highly specialized models which require only seconds or fractions of seconds of CDC 7600 execution time (or equivalent) per simulation). This conclusion is equally true for air quality model segments: For example, variances from a purely chemical model (or submodel), independent of other segments of an air quality model, can be reliably obtained only for cases in which about 10 or fewer chemical rates are sensitive parameters. We thus believe that policy-making applications will be best served by shifting the emphasis away from studying variances of limited scope and validity and, instead, to increased understanding of the physical and chemical cause-and-effect mechanisms with the aid of hybrid methods of sensitivity analysis.

Today's best approach to sensitivity analysis of complex air quality models would first decompose the subject model into a chemical component and a hydrodynamics component, noting several additional factors which apply to the model segmentation process:

(i) Segmenting is most effective when it is applied at the points of weakest mutual coupling in the air quality model. The chemical-hydrodynamics decoupling is an obvious first choice, based on present experience; improved decoupling points should always be sought.

(ii) Carefully considered application of model segmenting requires--and returns--greater understanding of the chemistry and physics in a given model.

(iii) If incorrect assumptions are made in model segmenting, quite often the result is that more--not less--is learned about both the air quality model and the simulated air quality problem.

Analysis of Sensitivities in the Chemical Component

The chemical mechanism of any large air quality model can be readily simulated by stand-alone automated chemical kinetics solvers such as those described in reference 17. Arbitrary conditions (e.g., initial conditions, species fluxes, dilution factors, and pollutant sources and sinks) can be abstracted from any spatial zone and time domain of the subject air quality model and provided as input for solution by the more efficient purely chemical kinetics solvers. This can be done repeatedly for representative conditions and circumstances in the original air quality model.*

Photochemical sensitivities can be determined most economically if rate parameters are at least piecewise constant in time (over one-hour intervals, for example) rather than instantaneously varying on a diurnal basis.

* This process of abstracting such conditions from a more general air quality model to a more specialized (photochemical) model is known as "linking". Linking has repeatedly been demonstrated to be effective in validating complex computational models in numerous scientific disciplines.

(Instantaneous diurnal variations exact such an extreme cost in kinetics integrations that all sensitivity methods can become unduly expensive.) Recognizing that the application of a kinetics sensitivity method is very much a matter of user preference, we will nevertheless indicate as a guiding suggestion that our present choice would be to use a direct variational method on the chemical component for several reasons:

- Economy of effort and computer cost. If decoupled from the kinetics solutions, a suitably automated direct sensitivity method is more economical than the other alternatives in terms of human effort and computer cost, particularly for new applications to large chemical mechanisms. Data handling is minimized as are arbitrary operational decisions such as trial and error of selection of rates, rate distributions, etc.
- Conceptual transparency. The measures $\delta c_i(\underline{x},t)$ of the time-honored method and $Z_{i,j}(t)$ of the direct method are both obtained continuously in time and are independent of statistical assumptions, unlike the measure $\langle Z_{i,j} \rangle$ of the FAST. The time-honored variable $\delta c_i(x,t)$ has an advantage of being valid to all orders. From experience with all of the alternative sensitivity methods (except for the

Green's function method which is not yet implemented for general use) we have usually found that the automated direct method gives the basic sensitivity information which is needed in a more organized and efficient, hands-off manner than the alternative methods. (Again, individual user preferences may very well differ from ours.)

- Potency. Direct variational methods come closest to fulfilling the full promise of systematic sensitivity analysis methods in chemical applications. No pre-screening (usually done by trial and error) is required in atmospheric applications of direct methods, whereas pre-screening is advisable for the FAST when more than about 20 parameters are to be tested. Pre-screening should always be minimized to the greatest possible extent because any oversights in this stage will compromise all subsequent evaluations. The measures $Z_{i,j}$ and $\delta c_i(\underline{x},t)$ of the direct and time-honored methods can also be used for best and worst-case identification, whereas $\langle Z_{i,j} \rangle$ from the FAST suppresses such information by the averaging process. By reverse reasoning, the FAST is the most effective method for providing averaged sensitivity coefficients in those special instances where the averages have meaning. It may also occur to some that the FAST can effectively compute $Z_{i,j}$, as well as $\langle Z_{i,j} \rangle$, by simply assigning zero uncertainties to all except the j^{-th} parameter. But we have seen that, in the absence of new advances, this apparent option of the FAST is grossly inefficient relative to the direct variational and time-honored alternatives, particularly for moderate to large-size chemical mechanisms.

- Ease of Application. A majority of practical users prefer the trial and error approach and tend to resist more formal sensitivity analysis methods which require their own numerical programs. The great intuitive appeal and immediacy of the time-honored approach ensure that it will continue as a favored sensitivity method. On the other hand, direct methods have now been developed and automated to a sufficient degree[7] [9] [11] for all types of chemical model use that widespread application should be near-at-hand. Koda et al[11] are similarly reducing the FAST method to semi-automatic general applicability in atmospheric chemistry problems.

The sensitivity testing of the purely chemical component is likely to be most meaningful if the systematic analysis (by the direct method) of rate and initial condition uncertainties is augmented by independent trial-and-error assessments of factors such as instantaneously varying photodissociation rates, more detailed characterizations of time dependent sources and sinks, and any other factors which were either by-passed or treated approximately by the direct method.

Analysis of Sensitivities in the Hydrodynamics Component

Whereas automated chemical kinetics solvers can be used efficiently to assess essentially unlimited chemical scenarios without running the full air quality model, the same is not true for the hydrodynamics component. The problem is that, even for non-reacting fluids, independent hydrodynamics solvers are not feasible for higher than one-dimensional transient models with arbitrary zoning, sources, sinks, and wind and diffusivity resolutions. Similarly, direct sensitivity methods have not yet been effectively automated for arbitrary scenario evaluations in higher than one-dimensional, transient hydrodynamics. It is therefore necessary for the

original air quality model, or some trivial derivative of it, to be used to generate necessary output data for sensitivity analyses of hydrodynamics factors. For example, the chemistry can be turned off by assigning zero rates to all chemical reactions in order to study the purely hydrodynamic (meteorological) characteristics of the air quality model.

With extensive basic understanding of the physical problem, the time-honored method would then be used almost exclusively to identify the key variables and their basic sensitivities to space and time dependent meteorological parameters, sources, sinks, initial conditions, and boundary conditions. The time-honored method would serve also as a means of pre-screening insensitive variables before applying the FAST to small groups of carefully selected hydrodynamics parameters. Correlated parameters (i.e. those which are represented by common functional relationships at all spatial zones) may also be treated effectively by the FAST in particular circumstances. Clearly, basic understanding of the physical system, coupled with sound intuition, is the most important element for incisive sampling and successful analysis of hydrodynamic sensitivities; for it is simply impossible to test every potentially sensitive parameter at every spatial grid point with <u>any</u> of the presently developed sensitivity methods.

NEEDED BREAKTHROUGHS IN SYSTEMATIC SENSITIVITY ANALYSIS METHODS

Breakthroughs which would provide truly major advances in sensitivity analysis applications fall into two major categories; first, there are <u>general advances</u> which would benefit <u>all</u> specific sensitivity and uncertainty analysis methods, and second, there are <u>specialized advances</u> which will benefit, exclusively, a particular sensitivity method.

General Advances

- Model execution speed. We have seen that several sensitivity methods would become more attractive if the cost of air quality simulations were on the order of 1/100 cents per run. However, this possibility appears to be out of reach for even perfectly optimized 2-D and 3-D transient air quality models with envisioned computers of the next generation or two.

- Advances in partial differential equations. Significant advances in PDE numerical analysis are likely over the next 5-10 years which may dramatically improve air quality modeling. First, numerical integration of non-linear systems of PDE's is in the midst of major advances. Finite element methods with adaptive gridding features[19] have the capacity to use far fewer grid points, located at optimal positions. These features tend to eliminate numerical diffusion and the overshooting errors which have always plagued the finite difference methods which are used in current-generation models. These new approaches are, additionally, very amenable to automatic compilation of arbitrary problem scenarios in higher dimensions, in a manner which is very much akin to the automated kinetics (ODE) solvers which were mentioned in previous sections. These general advances undoubtedly will be many years in coming to fruition, unless special initiatives are undertaken.

- Correlated variables allow significant reductions in execution costs for all systematic sensitivity analysis methods. The identification of correlations in winds and diffusivities, for example, will most likely come from very fundamental considerations of fluid dynamics. Such studies are far more basic and rigorous than most air quality models, per se, and will thus require many more years of highly detailed theoretical and experimental analysis.

Specialized Advances

- The FAST method would be markedly improved by more efficient parameter sampling which relates to the selection of incommensurate Fourier frequencies.
- The FAST method would run markedly faster if correlated parameters can be identified and represented by a common parametric variation, which in turn reduces the required number of independent sample points.
- The direct variational methods are markedly improved when the sensitivity computations are decoupled from the kinetics or hydrodynamics computations. A very modest amount of research (about 1 person-year) will now accomplish the development of reliable decoupling methods.
- The direct variational methods will advance markedly when 2-D hydrodynamics models can be automatically compiled for arbitrary problem scenarios and solved by generalized hands-off PDE solvers (such as is presently the case with automatic kinetics solvers which use the Gear ODE solution method[17] [8]). Additionally, the sensitivity equations must be amenable to the same measures. At Science Applications, Inc., in Pleasanton, California, in-house efforts are, in fact, remarkably near (within 3 person-years) of accomplishing this advance using very potent finite element methods. (We are aware of only one other such effort, in a different field, which is near a comparable point of advancement, so it is difficult to give a more universal status estimate than the one above which is based on our own work.)
- Finally the Green's function method may ultimately offer some advantage over alternative sensitivity methods for purely chemical systems. It is still too early to tell. (We estimate that 1-2 person-years may yet be required to satisfactorily implement this method in a suitable automatic format for arbitrary mechanism evaluations.)

REFERENCES

1. Reynolds, S.D., P.M Roth and J.H. Seinfeld, "Mathematical Modeling of Photochemical Air Pollution -I. Formulation of the Model.", Atmos. Environ., 7, 1033-1061. (1973).

2. Mac Cracken, M.C., D.J. Wuebbles, J.J. Walton, W.H. Duewer, and K.E. Grant, "The Livermore Regional Air Quality Model: I and II", J.A.M., 17, No. 3, 254 (March 1978).

3. Sklarew, R.C., M.A. Joncich, K.T. Tran, "An Operational Air Quality Modeling System for Photochemical Smog in San Diego Air Basin", Fourth Symposium on Turbulence, Diffusion, and Air Pollution, 301, (AMS Jan 15-18, 1979).

4. Hecht, T.A., J.H. Seinfeld, and M.C. Dodge, "Further Development of a Generalized Kinetic Mechanism for Photochemical Smog", Environ. Sci. Technol., Vol. 8, No. 4, pp. 327-339 (1974).

5. Whitten, G.Z. and H. Hogo, "Mathematical Modeling of Simulated Photochemical Smog", EPA Report EPA-600/3-77-011 (Jan. 1977).

6. Gelinas, R.J. and Skewes-Cox, P.D., "Tropospheric Photochemical Mechanisms", J. Phys. Chem., 81, 2468 (1977).

7. Dickinson, R.P. and Gelinas, R.J., "Sensitivity Analysis of Ordinary Differential Equation Systems", Journal of Computational Physics, Vol. 21, No. 2, 123-143, (June 1976).

8. Hindmarsh, A.C. and G.D. Byrne, "EPISODE: An Experimental Package for the Integration of Systems of Ordinary Differential Equation Systems", UCID-30112, Lawrence Livermore Laboratory (May, 1975).

9. Atherton, R.W., R.B. Schainker, and E.R. Ducot, "On the Statistical Sensitivity Analysis of Models for Chemical Kinetics", A.I.C h.E. J., 21, 441 (1975).

10. Gelinas, R.J., "Diurnal Kinetic Modeling", Proceedings of the International Conference on Structure, Composition and General Circulation of the Upper and Lower Atmosphere and Possible Anthropogenic Perturbations, Vol. II, Jan. 14-25, 1974, IAMAP/IAPSO, Melbourne, Australia, (1974).

11. Koda, M., A H. Dogru, and J.H. Seinfeld, "Sensitivity Analysis of Partial Differential Equations with Application to Reaction and Diffusion Processes", preprint (1978).

12. Walter, W., Differential and Integral Inequalities, (Springer-Verlag, New York, 1970).

13. Porter, W.A., Int. J. Control, 5, 393 (1967).

14. Hwang, J.T., E.P. Dougherty, S. Rabitz, and H. Rabitz, "The Green's Function Method of Sensitivity Analysis in Chemical Kinetics", preprint (1978).

15. Stolarski, R.S., D.M. Butler, and R.D. Rundel, "Uncertainty Propagation in a Stratospheric Model, 2. Monte Carlo Analysis of Imprecisions due to Reaction Rates", JGR, in press (1978).

16. Cukier, R.I., C.M. Fortuin, K.K. Schuler, A.G. Petschek, and J.H. Schaibly, J. Chem. Phys. 59, 3873, (1973).

17. Dickinson, R.P. and Gelinas, R.J., "SETKIN", in L. Lapidus and Schiesser (eds.), Numerical Methods for Differential Systems, (Academic Press, 1976), pp 167-180.

18. Penner, R.C., private communication, also see A.A. Boni and R.C. Penner, Combust. Sci. Technol. 15, 99, (1976).

19. Doss, S., private communication (1978).